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URANIUM ENRICHMENT

MARTIN MARIETTA

TECHNICAL INFORMATION FOR ASSESSING COST RESPONSIBILITY FOR DECONTAMINATION AND DECOMMISSIONING OF THE GASEOUS DIFFUSION PLANTS

February 1992

MANAGED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

UEO-216 (Draft)

TECHNICAL INFORMATION

FOR

ASSESSING COST RESPONSIBILITY FOR

DECONTAMINATION AND DECOMMISSIONING

OF THE

GASEOUS DIFFUSION PLANTS

H. M. NORITAKE, OAK RIDGE

J. H. THOMAS, PADUCAH

E. R. WAGNER, PORTSMOUTH

February 1992

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EXECUTIVE SUMMARY

INTRODUCTION

The cost for the eventual decontamination and decommissioning (D&D) of Department of Energy (DOE) gaseous diffusion plants (GDP) has been estimated in recent studies to exceed $15 billion. The three diffusion plants at Oak Ridge, Paducah, and Portsmouth were operated exclusively for defense and government purposes for the first 15 to 20 years before they were gradually shifted to providing enrichment services for the commercial sector to fuel nuclear power plants. The DOE plants are now operated almost exclusively for commercial customers. Thus, the assignment of cost responsibility between defense and commercial customers is an important element of planning and implementing D&D. A few facilities such as the toll enrichment facilities at each site and the feed plant at Portsmouth were built solely to meet the needs of commercial operation. These few facilities have been found to contribute only a small fraction[1] to the estimated cost of D&D of all of the buildings at the three GDP sites. Buildings and equipment that produce highly enriched uranium at Oak Ridge and Portsmouth have been used in support of defense and government research programs. All other buildings have been operated to meet the demands of both the defense and commercial customers.

The cost of D&D of the gaseous diffusion plants and associated auxiliary facilities is increased manyfold relative to non-nuclear facilities by the fact that the key material being processed is uranium, which is mildly radioactive with an extremely long half-life and with daughter products that are also radioactive. The assignment of liability for D&D depends on the relative responsibility of both government and commercial programs for creating the need for eventual D&D of the GDPs. The need for D&D was created by the exposure to slightly radioactive process gas; the effort required to perform D&D is a function not only of the level of contamination but also of the extent of contamination as measured by the volume of material[2] to be ultimately disposed of as low-level waste (LLW). Early records providing details on the levels and extent of radioactive contamination as a function of time were found to be unavailable, so other sources of information were investigated.

This study was conducted to locate historical information that could provide a technical basis for determining responsibility for contamination of the enrichment facilities and equipment. Seven areas of information are documented regarding the occurrence and extent of contamination at the three GDP sites. Three of the areas address the internal contamination of the process equipment. These include the following:

- Inventory difference (ID). These data represent the amount of uranium fed to the cascade that did not emerge in either the product or tails streams and is considered to be retained within the cascade.

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1. The D&D costs of these three facilities constitute only 0.02% of the total cost for the GDPs (based on Ebasco's "Cost Estimate for D&D of the GDPs," September, 1991)

2. The additional D&D cost incurred due to the slightly larger process equipment installed during the CIP/CUP program is extremely small, being approximately 0.1% of the total cost of D&D.

- o Nondestructive assay (NDA) surveys of process equipment. These on-going surveys are locating the presence of uranium deposits within the process equipment.

- o Reprocessed uranium (RU). Feeding of reprocessed uranium from defense reactors introduced radioactive transuranic (TRU) material into the GDPs.

Four of the areas are concerned with the general contamination of the interior of the process buildings and auxiliary buildings in which uranium was processed. These include the following:

- o Equipment exchanges. Replacing process equipment involves opening the gaseous diffusion stages to the atmosphere in order to perform the exchange. A small amount of process gas is unavoidably released during each such operation.

- o Process gas releases. Accidental releases of process gas were found to occur in the process buildings as well as in other buildings where UF_6 is handled. These releases result in radioactive contamination of the areas where they occur.

- o Contamination within process buildings. Data from some surveys within the process buildings are available to indicate the current extent of contamination.

- o Urinalysis data. Urinalysis data have been collected on personnel considered to have been exposed to UF_6 environment. The number of such exposures is considered to be representative of the extent of radioactive contamination occurring on the GDP sites.

INTERNAL CONTAMINATION

Uranium Deposition and Inventory Difference (ID) Data

The chemistry and surface physics of uranium deposition on internal equipment surfaces in the UF_6 process gas environment are well understood. The four principal mechanisms for internal deposition, chemisorption, physical adsorption, corrosion, and hydrolysis, have been evaluated not only in laboratory studies but also by plant experience for a spectrum of structural materials of the cascade including nickel, aluminum, nickel-plated steel, copper, and steel. The analytical model for uranium deposition shows that the total uranium deposition in the commercial period is below the levels of the 1960s. Historical Inventory Difference data from nuclear material accountability records at Paducah and Oak Ridge support this analytical model. The Portsmouth plant is unique because of factors related to high-assay production; nonetheless, the Portsmouth accountability data also support the fact that deposition rates in improved equipment are substantially lower than rates in the original process equipment.

Nondestructive Assay (NDA) of Process Equipment

Surveys of process equipment are currently being conducted in the shutdown production equipment at ORGDP to locate residual deposits that may present a criticality concern during eventual D&D. Nondestructive assays (NDAs) of highly enriched uranium (HEU) Buildings, K-25 and K-27, have been completed, and preliminary surveys have been performed in Buildings K-29 and K-31. About half of the process equipment in K-31 was changed out during the Cascade Improvement and

Cascade Uprating Programs (CIP/CUP) in 1975-1982, while the other half represents equipment that was installed during the first Improvement Program, 1957-1962. The preliminary data from the survey reveal that the number and size of uranium deposits in the "old" equipment (circa 1957) are much greater than in the "newer" equipment, which is in keeping with the improvement in equipment and operating procedures.

Feeding of Reprocessed Uranium

Between 1953 and 1976, reprocessed uranium from plutonium production reactors at Hanford and Savannah River was fed to the gaseous diffusion plants as part of the defense program. This material contained transuranic and fission product radionuclides. While most of the material was introduced at the Paducah plant, the radioactive contaminants spread over a period of years to all three plants. Transuranic elements (particularly neptunium) and the fission product, ^{99}Tc, can now be detected in most of the process equipment and to a lesser extent in general building contamination. This additional contamination of equipment and process buildings is caused almost entirely (99%) by the defense program decision to feed reprocessed uranium to the diffusion plants.

CONTAMINATION OF BUILDING INTERIORS

Equipment Exchanges

During the maintenance of equipment failures and replacement of obsolete hardware, uranium contaminants were released from within equipment to the interior of process buildings. Particularly during the early years, failures were quite numerous as new designs were rushed into production to meet urgent material requirements for the weapons program. In addition, an initial Improvement Program was undertaken in 1957-1962 in which a large percentage of process equipment was replaced with improved equipment to enhance performance. These exchanges have been tabulated to indicate the number of opportunities for contamination of the cell and operating floors of process and maintenance buildings. Some 8,000 converters and compressor exchanges at Oak Ridge, almost 9,000 exchanges at Paducah, and almost 9,000 at Portsmouth (exclusive of even greater numbers of compressor seal changes) have been made since the start of operation. A major portion of these occurred in the years before 1970 when defense production was the primary mission. Although historical floor contamination records are lacking, interviews conducted with personnel in building operations, maintenance, and health physics confirm that contamination of process buildings occurred each time that process lines were cut to exchange equipment. Improvements in contamination control practices in recent years have ended the spread of contamination within process buildings. As cleanup actions are undertaken to meet DOE Order 5480.11, building contamination levels in operating plants are expected to be reduced over the remaining life of the facilities.

Record of Process Gas Releases

Accidental releases of process gas in process buildings and auxiliary buildings are another significant source of general building contamination. Records of major releases (those releasing 1 kg of uranium or more) have been maintained by the Nuclear Material Accountability departments for the history of each site. Accidental releases typically in the form of gaseous UF_6 result in contamination of the immediate area of the release and vicinity depending upon circumstances. A few larger releases have spread contamination over a significant portion of some process buildings. A study of these releases,

many occurring in auxiliary buildings such as the feed facility, indicates that the great majority of the larger releases occurred in the 1950s and 1960s during initial years of operations.

Contamination Surveys

Most recently, in July 1991, surveys of floor contamination in process buildings were conducted at Oak Ridge, Paducah, and Portsmouth. Beta-gamma contamination was found to some extent in every building, indicating scattered but widespread contamination of uranium and its daughter products. Only at Oak Ridge where the K-25 and K-27 buildings that had been used only for high-assay production and that had been shut down in 1964 could a distinction be made between defense-related and commercial-related facilities. The instrument readings obtained in K-25 and K-27 showed about the same extent of contamination as was found in the low-assay facilities, K-31 and K-33, which operated until 1985. More recent health physics and radiation protection procedures regarding the designation and access to contamination zones have reduced the spread of contamination to an extent that many areas are now regarded by plant health physics personnel as being less contaminated than before the implementation of these policies.

Improvements in contamination control practices include (1) posting regulated and contaminated areas, (2) requiring special contamination clothing and shoe protection in those areas with changeout before exiting a contamination control zone, and (3) monitoring stations and procedures for monitoring before exiting controlled areas. Current procedures regarding equipment removals require (1) purging of cascade cells to a concentration of less than 2 ppm (versus past practice of 10 ppm) before opening for equipment maintenance, (2) covering openings and seal with tape, and (3) using gaskets and clamps over metal covers. Significant improvements have been made in the capability of instruments to detect lower concentrations of uranium in gas streams, thus allowing requirements for amounts of uranium in exhaust gases to be lowered.

Urinalysis as a Measure of Contamination Level

Bioassay, specifically, urinalysis, has been a routine part of the health physics and radiation protection program at the three sites because of the predominance of soluble uranium compounds. Complete historical data on the urinalyses of personnel engaged in particular activities or working in certain areas was assembled for both the Oak Ridge and Portsmouth plants. While not pinpointing the particular activities or locations that led to the need for urinalysis sampling, the data provide a record that demonstrates personnel were exposed to a uranium-contaminated workplace. The data also show very clearly that the preponderance of such exposures occurred in the years before 1970, with a significant increase during the years of the first Improvement Program in 1957-1962. This again was the period of production solely for the defense program. Urinalysis data are only an indirect measure of historical contamination because of dependence not only upon the extent of contamination present but also upon the level of precautionary measures taken. Current practice involves much closer monitoring of the air and surfaces of the work areas and more protective gear being worn. Consequently, ingestion of uranium is much less likely in recent years than during the fifties and sixties when the rules regarding personnel protection against contamination in the workplace were much less stringent. Thus, urinalysis must be regarded as a measure of both the extent of contamination as well as the extent of personnel protection being practiced at the time. Allowable standards in both areas have become more stringent with the passage of time.

OBSERVATIONS AND CONCLUSIONS

Information presented in this report provides a database from which preliminary conclusions can be drawn regarding responsibility for the cost of D&D of the GDPs at Oak Ridge, Paducah and Portsmouth. The requirement for decontamination or disposal of process equipment as low-level waste because of internal radioactive contamination was established by early operation of the equipment for the defense mission.

- Internal contamination of process equipment occurs when process gas is first introduced because of immediate chemisorption of uranium fluorides.

- The NDA survey confirms the fact that equipment installed in the fifties and sixties contains greater uranium deposits than new equipment installed in the late seventies. This fact is also indirectly supported by the inventory difference data for each plant.

- Introduction of reprocessed uranium from defense production reactors into the gaseous diffusion process contaminated all three plants with trace amounts of transuranics and ^{99}Tc. Many large buildings at the GDP sites have become contaminated with radioactivity during the many years of operation and represent a very large D&D cost category. Building contamination is dependent on sources of uranium released within the process buildings. The data for two principal mechanisms for release, accidental releases and equipment changeouts, show that the early operation of the facilities was characterized by frequent maintenance actions and significant accidental releases. Consequently, by the early 1960s, process buildings and other buildings were significantly contaminated.

- A large number of equipment exchanges to replace failed equipment or to install upgraded equipment have provided opportunities to spread contamination within process buildings. Many of these changeouts occurred before 1970, and contamination controls have been significantly enhanced since the early years of operation.

- Process gas releases are another source of contamination of buildings on the sites. A majority of these releases occurred during the 1950s and early 1960s.

- Surveys of contamination within process buildings have proved that (1) contamination exists to some extent in every process building; and (2) K-25 and K-27 buildings at Oak Ridge that were shut down in 1964 have contamination levels comparable to or higher than the low-assay process buildings, K-31 or K-33 (K-29 was subjected to a major release), indicating that incremental contamination since 1965 has been quite small.

- Urinalysis samples of workers confirm the fact that many workers were exposed to uranium released in their work areas in the early years of operation and during programs for equipment exchanges.

Based on the information given, the level of decontamination effort required for process buildings is no greater now than the effort that would have been required in 1965 if all the facilities had been required to have been decommissioned at that time.

While no single set of data provides a clear basis for allocation of D&D liability, the preponderance of all data reviewed clearly supports the position that a large percentage of D&D costs are the responsibility of the U. S. Government. The gaseous diffusion plants were originally constructed to fulfill the government's need for enriched uranium. Only after the first two decades of government use did the commercial sector derive any benefit from the existence of these plants. In 1969, when commercial production began, the diffusion plants were already contaminated and the need to conduct D&D was well established. Contamination with radioactivity at the three GDP sites consists of internal contamination of equipment and contamination of building interiors. As suggested by historical data gathered to date, the introduction of commercial production did not add greatly to the existing overall level of contamination. This is particularly true of internal contamination of equipment where some data suggest that the levels have actually been reduced during the years of commercial production. Furthermore, the large volume of contaminated material has remained relatively unchanged with only modest increases associated with process equipment upgrades.

INTRODUCTION

Cost responsibility for decontamination and decommissioning (D&D) of the gaseous diffusion plants (GDP) can be segregated into government liability and commercial liability. Certain facilities at gaseous diffusion plant sites have been used exclusively for furnishing uranium materials to government programs, primarily for national defense and research uses. These facilities include the K-25 and K-27 buildings at Oak Ridge, the feed and metal plants at Paducah, and the X-326 cascade building and the X-345 vault at Portsmouth. Costs for D&D of these facilities are clearly a responsibility of the government. On the other hand, a few facilities were built in the late 1960s and early 1970s exclusively for the commercial program. These are the toll enrichment facilities at each plant and the UF_6 feed facility at Portsmouth[3]. The costs of D&D for these facilities should therefore be an obligation of the commercial program.

Other facilities at the three sites were built for government defense programs in the 1940s and 1950s and were used exclusively until the 1960s for these programs. In the 1960s, a small portion of deliveries (less than 5%) was used for commercial nuclear power. The commercial toll enrichment program started officially in 1969. Through 1991, commercial customers have received 55% of total SWU deliveries from gaseous diffusion plants. Today and in the future, commercial deliveries will represent in excess of 95% of total production.

The cost of D&D of the gaseous diffusion plants and associated auxiliary facilities is increased manyfold relative to nonnuclear facilities by the fact that the key material being processed is uranium, which is mildly radioactive (alpha emission of 2.52×10^4 dps/gm at 4.76 Mev) but has an extremely long half-life of 4.5×10^7 years and has daughter products that are also radioactive. Decontamination of large surface areas and disposal of large quantities of diffusion plant material and equipment as low-level waste (LLW) with its attendant rules and restrictions are a costly operation when compared to disposing of a facility with no radioactivity involved.

The assignment of commercial liability for D&D depends on the relative responsibility of both government and commercial programs for generating the need for eventual decontamination and decommissioning of the GDPs. An earlier DOE cost allocation between government and commercial programs was based on the assumption that the cleanup liability was directly proportional to the number of SWUs delivered. This approach did not have a technical basis, but appears to have been a convenient method of splitting the cost based on an approximate measure of relative benefits.

To develop a technical basis for assigning cost responsibility, a survey of historical records as well as a technical analysis of the deposition of radioactive contaminants was initiated. Information on historical building contamination measurements at Oak Ridge, Paducah and Portsmouth and a review of technical information on internal contamination have increased our understanding of the time-dependent development of contamination patterns within the diffusion plants. Only a few records directly provide a measure of the historical evolution of either internal or external contamination. An excellent technical basis for understanding the contamination of process equipment by UF_6 exists, however; and this technical base is supported by accountability data, experimental measurements, and

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3. These three facilities constitute only 0.02% of the total D&D cost (based on Ebasco report, *Cost Estimate for D&D of the GDPs*, September 1991)

reset nondestructive assay measurements of shutdown equipment. Also, numerous historical records relate to the release of uranium that caused building contamination. This report summarizes these technical evaluations and historical records that relate directly or indirectly to the generation of contamination in equipment and buildings at the three gaseous diffusion plants.

TECHNICAL BASIS FOR INTERNAL CONTAMINATION

INVENTORY DIFFERENCE DATA[4]

Mechanisms of Uranium Deposition

Chemisorption. Radioactive material is deposited in a gaseous diffusion cascade by three different mechanisms, i.e., chemisorption, metal corrosion and hydrolysis. An initial deposition of uranium occurs almost instantaneously because of chemisorption of uranium hexafluoride (UF_6) on equipment surfaces. Another feature of chemisorption is that it occurs everywhere that the gas can reach, including the interior of porous materials and deep, closed-end recesses. Since pressures and temperatures required to remove this chemisorbed UF_6 cannot be achieved in cascade equipment, all of the chemisorbed gas will remain on the cascade surfaces after in-place cleanup of equipment following shutdown. The quantity of chemisorbed material does not change with exposure time; it is always present following initial exposure. Because of the essentially instantaneous nature of UF_6 chemisorption, the need for decontamination of the gaseous diffusion plants was undoubtedly established as soon as the equipment had been exposed to UF_6 for the first time. By the same token, the volume of equipment to be disposed as LLW was established at the same time except for the small increase in volume created by the slightly larger equipment installed during the CIP/CUP program undertaken in the late seventies.[5]

Physical Adsorption. A relatively small additional amount of physically adsorbed UF_6 will be present on the surfaces while equipment is in operation, but this physically adsorbed material is loosely bound to surfaces and is completely removed along with gaseous material during normal cascade shutdown, evacuation, and purging procedure. The quantity of physically adsorbed UF_6 accounts for about 10% of the total adsorbed material; the other 90% is the very tightly bound chemisorbed material.

Corrosion. The internal structural materials of the cascade, which are principally nickel, aluminum, nickel-plated steel, copper, and small quantities of iron, are corroded by UF_6, producing a deposit of solid, reduced uranium fluoride. The rate of this deposition is minimized in diffusion cascades by prestabilization of the metal surfaces with fluorine. Since a protective film is not formed on iron surfaces, prestabilization is not effective in reducing the subsequent corrosion rate of iron surfaces.

Equipment initially installed in the diffusion plants exhibited much higher rates of reaction with

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[4] This section prepared by R. L. Ritter, Enrichment Technology Operations Division, Oak Ridge.

[5]. The slight increase in volume of waste due to the CIP/CUP program is estimated as increasing the total D&D cost by approximately 0.1%.

process gas than did equipment installed at later dates, and therefore higher rates of deposition of solid uranium-containing deposits were observed during early periods of cascade operation.

Hydrolysis. Solid material that contains uranium can also be deposited on cascade surfaces by hydrolysis of UF_6 when water vapor enters the cascade through small leaks in process equipment. The rate of deposition as a result of this mechanism is extremely difficult to quantify, since the rate is dependent on the frequency and extent of wet air inleakage, which can vary widely with time and with cascade location. Most of the solid uranium material formed by this mechanism is deposited on surfaces in the vicinity of the inleakage point. (Areas of diffusion cascades where uranium contamination continues to accumulate are in deep recesses that are closed at one end and that contain exposed, unplated steel surfaces.)

Inventory Difference Data

A monthly uranium inventory is conducted at each of the diffusion plants during which the quantity of uranium present in the gas phase as UF_6 is calculated. From these results and the net of feed and withdrawals to the cascade, an inventory difference (ID) is determined for both total uranium and ^{235}U mass balances for the period. ID data can provide important information concerning the retention of some of the process gas as an adsorbed film or as UO_2F_2 deposits.

The cumulative ID for the Oak Ridge cascade during the period from October 1964 at which time the K-25 and K-27 buildings were removed from service through August 1984 is shown in Table 1 and plotted in Figure 1. Two important points can be made from these data. First, a significant reduction in the cumulative ID occurred during 1975, with the ID decreasing from about 27,000 KgU at the beginning of 1975 to about 18,500 kgU at the beginning of 1976, representing a recovery of more than 30% of the total ID accumulated since 1964. This is attributed to cleanup of portions of the deposits of uranyl fluoride formed during preceding years. Secondly, a much-reduced rate of ID accumulation is seen for the later years of cascade operation, as indicated by the nearly horizontal line in the years beyond 1977. These two effects combined to produce, by 1984, a reduction in the contamination in process equipment as compared to the contamination in earlier years.

Table 2 is a summary of the plant cumulative ID data from Paducah[6], which is shown plotted in Figure 2. The much larger value for the Paducah ID as compared to the Oak Ridge ID data of Figure 1 is primarily the result of the different starting dates; the earlier years employed in the Paducah data include relatively large IDs associated with feed plant operation, whereas operation of the Oak Ridge feed plant was terminated before 1964 and therefore ID associated with this source is not included in the Oak Ridge data of Figure 1. The rapid increase in ID between 1953 and 1969 is quite similar to the rapid increase between 1965 and 1974 shown in Figure 1. A decrease in ID occurs between 1970 and 1974 followed by a much slower rate of increase after 1976, a pattern very similar to that found at Oak Ridge. Although the results are not so clear-cut as at Oak Ridge, apparently the cascade equipment is no more contaminated at the end of 1989 than it was at the end of 1969.

6. G. W. Tuttle, *Discussion of Historical Inventory Difference (ID) at the Paducah GDP, FY 1953 through FY 1989, November 1990*, (KY/D-3998, Rev. 1)

TABLE 1. OAK RIDGE INVENTORY DIFFERENCE
1965 – 1984

YEAR	TOTAL PLANT AID, KGU	
	ANNUAL AID	CUMULATIVE AID
1965	2,617	2,617
1966	5,431	8,048
1967	1,175	9,223
1968	(778)	8,445
1969	9,531	17,976
1970	2,403	20,379
1971	130	20,509
1972	3,857	24,366
1973	2,344	26,710
1974	(578)	26,132
1975	(3,410)	22,722
1976	(7,834)	14,888
1977	(410)	14,478
1978	5,699	20,177
1979	(7,493)	12,684
1980	347	13,031
1981	5,038	18,069
1982	1,112	19,181
1983	331	19,512
1984	(2,987)	16,525

FIGURE 1. URANIUM INVENTORY DIFFERENCE - OAK RIDGE CUMULATIVE TOTALS, 1965 - 1984

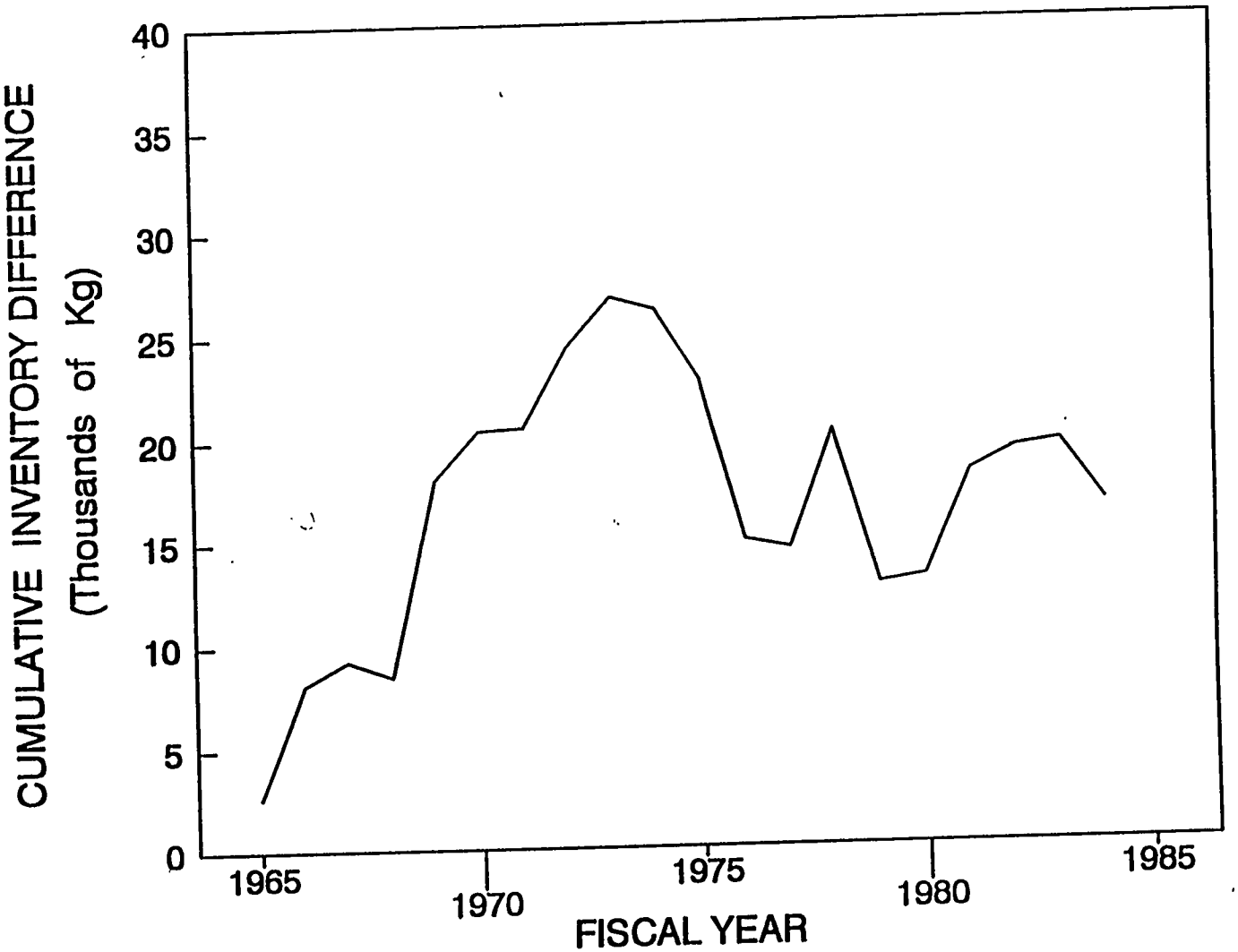


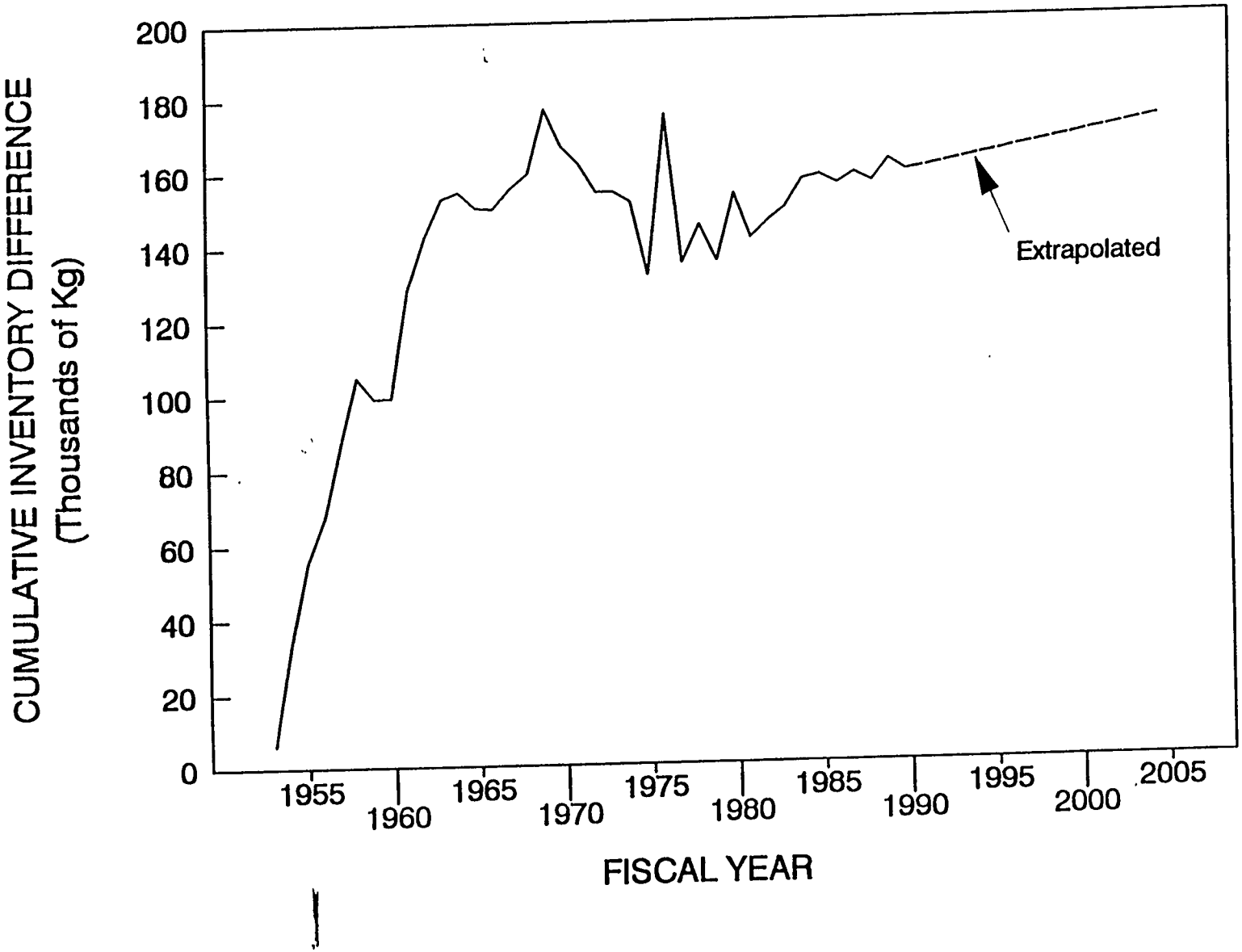
TABLE 2. PADUCAH INVENTORY DIFFERENCE
1953 – 2005

YEAR	TOTAL PLANT AID, KgU	
	ANNUAL AID	CUMULATIVE AID
1953	8,256	8,256
1954	27,480	35,736
1955	21,594	57,330
1956	12,413	69,743
1957	19,653	89,396
1958	17,649	107,045
1959	(5,845)	101,200
1960	131	101,331
1961	28,902	130,233
1962	14,231	144,464
1963	10,263	154,727
1964	1,806	156,533
1965	(4,379)	152,154
1966	(287)	151,867
1967	5,419	157,286
1968	4,024	161,310
1969	17,538	178,848
1970	(10,349)	168,499
1971	(4,873)	163,626
1972	(7,593)	156,033
1973	96	156,129
1974	(3,011)	153,118
1975	(19,490)	133,628
1976	43,431	177,059
1977	(40,330)	136,729
1978	10,165	146,894
1979	(9,652)	137,242
1980	17,858	155,100
1981	(12,052)	143,048
1982	4,378	147,426

TABLE 2. PADUCAH INVENTORY DIFFERENCE (cont.)
1953 – 2005

YEAR	TOTAL PLANT AID, KgU	
	ANNUAL AID	CUMULATIVE AID
1983	3,726	151,152
1984	7,597	158,749
1985	1,061	159,810
1986	(2,425)	157,385
1987	2,839	160,224
1988	(2,464)	157,760
1989	5909	163,669
1990	−2942	160,727
1991	975	161,702
1992	975	162,677
1993	976	163,653
1994	975	164,628
1995	975	165,603
1996	975	166,578
1997	976	167,554
1998	975	168,529
1999	975	169,504
2000	975	170,479
2001	976	171,455
2002	975	172,430
2003	975	173,405
2004	976	174,381
2005	975	175,356

FIGURE 2. URANIUM INVENTORY DIFFERENCE - PADUCAH CUMULATIVE TOTALS, 1953 - 2005



While the absolute values of ID are quite different between Figures 1 and 2, the shapes of the curves including the rapid rise in the early years followed by a reduction of ID in the early to mid-seventies are very similar. After about 1976, the increase in ID is very small at Oak Ridge while a reduced but still noticeable increase is evident at Paducah. A very definite reduction in ID occurs at Oak Ridge by 1984; however, at Paducah, the reduction obtained in the mid-seventies is almost erased by 1989.

ID data for Portsmouth[7] are shown in Table 3 and are plotted as cumulative ID in Figure 3. The rapid increase in ID in the early years followed by a noticeable reduction in the rate is in general agreement with the other plants.

While marked decreases have been observed in the rate of retained inventory (RI) accumulation, there will still be a small but measurable accumulation of cascade RI with continued operation of the cascades. Probably the best method for estimating the future accumulations of plant ID is to extrapolate the data from recent years in a straight-line fashion, which has been done; and the results are also shown in Figures 2 and 3 for the Paducah and Portsmouth cascades respectively. Straight-line least square fits were made to the cumulative ID data for each plant site for the period 1974-1989 and extrapolated to the year 2005. The results indicate ID accumulation rates of 975 and 649 kgU/yr for the Paducah and Portsmouth plants, respectively. No estimate of the effect of shutdown of the HEU portion of the Portsmouth cascade has been made, because determining the portion of the plant ID associated with this equipment is not possible at this time.

The above estimate of the plant IDs in future years is an estimate of the total site ID. The calculational model developed in the early 1980s has been used to provide an estimate of the rate of ID accumulation in the 00 and 000 size cascade equipment; increases of 0.115 and 0.163 KgU/yr/stage are estimated for 000 and 00 equipment, respectively. Using these estimates, an additional accumulation of cascade retained inventory of 3,840 kg U at Paducah and 2,560 kg U at Portsmouth is expected to occur during the 16-year period from 1989 to 2005, equivalent to accumulation rates of 240 and 160 kgU/yr for the Paducah and Portsmouth sites, respectively. These results indicate that about 25% of the total plant ID accumulation in the future will occur internally in the cascade equipment. Note that this estimate includes only internal deposits in the cascade equipment and makes no allowance for U losses in the other material balance areas such as, for example, decontamination. The total cascade RI accumulation will undoubtedly decrease at Portsmouth when the X-326 equipment is placed in standby, but this should not affect the accumulation rate in the equipment remaining on-stream.

Calculation of the Cascade RI

Estimates of the cascade RI, defined as that portion of the uranium ID that is retained in the cascade equipment in the form of either solid deposits or as adsorbed UF_6, have been made for many years. The early estimates were based on data obtained during decontamination of cascade equipment, in which the quantity of uranium recovered during the decontamination process was tabulated as a function of on-stream service time. From such data, factors representing the rate of accumulation of RI with time were derived for the various pieces of process equipment, and the RI was estimated by simply multiplying the cascade on-stream time by the appropriate factors. These factors were last

7. D. A. Shisler, Letter to H.M. Noritake, Portsmouth Gaseous Diffusion Plant Historical Cascade Inventory Difference, December 13, 1991

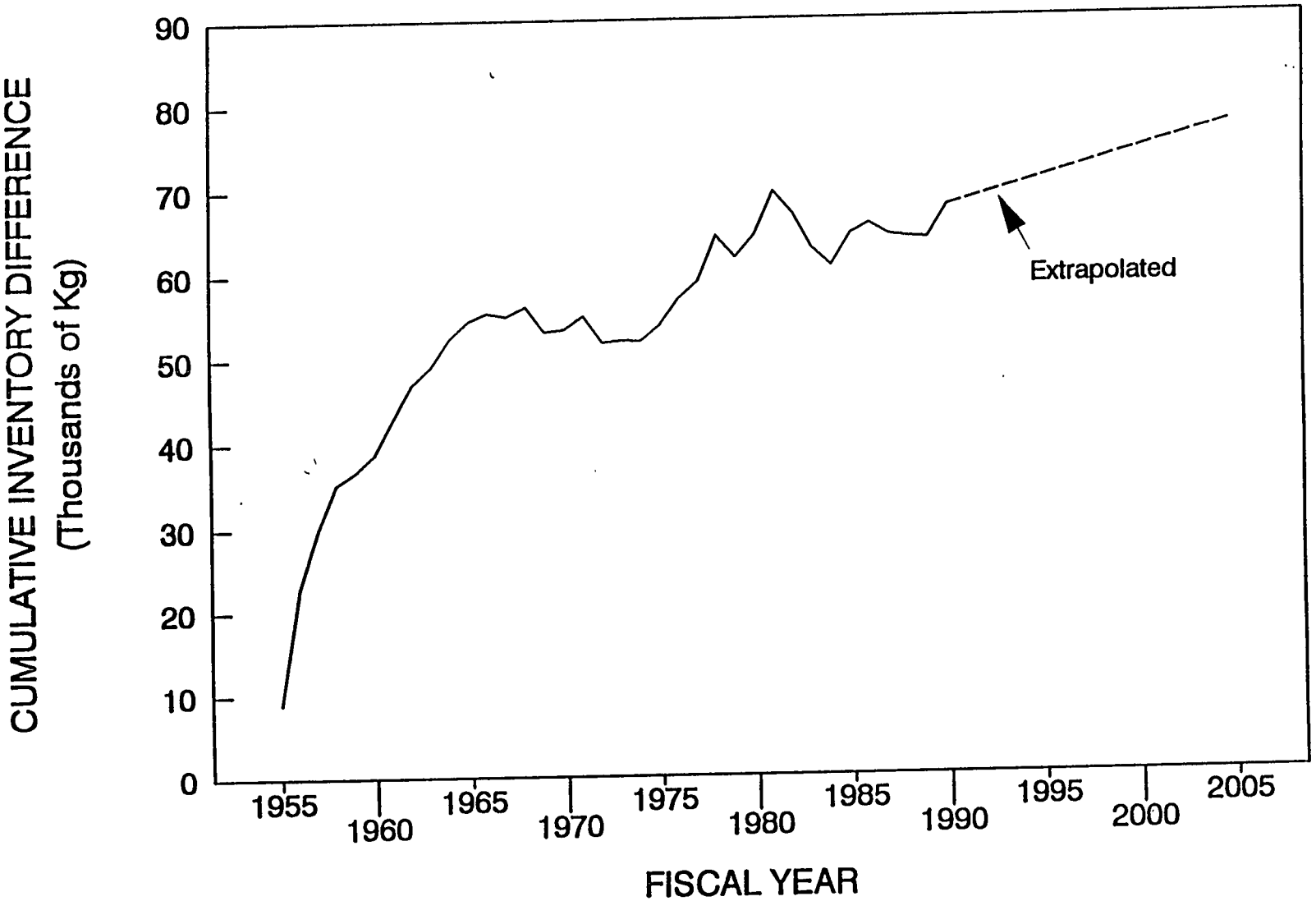
TABLE 3. PORTSMOUTH INVENTORY DIFFERENCE
1955 – 2005

YEAR	TOTAL PLANT AID, KGU	
	ANNUAL AID	CUMULATIVE AID
1955	8,989	8,989
1956	13,955	22,944
1957	7,079	30,023
1958	5,130	35,153
1959	1,517	36,670
1960	1,967	38,637
1961	4,141	42,778
1962	4,160	46,938
1963	2,092	49,030
1964	3,325	52,355
1965	2,100	54,455
1966	915	55,370
1967	(447)	54,923
1968	1,115	56,038
1969	(2,965)	53,073
1970	298	53,371
1971	1,533	54,904
1972	(3,131)	51,773
1973	180	51,953
1974	(54)	51,899
1975	1,779	53,678
1976	3,121	56,799
1977	2,083	58,882
1978	5,420	64,302
1979	(2,619)	61,683
1980	2,673	64,356
1981	5,142	69,498
1982	(2,715)	66,783
1983	(4,053)	62,730
1984	(2,112)	60,618

TABLE 3. PORTSMOUTH INVENTORY DIFFERENCE (cont.)
1955 – 2005

YEAR	TOTAL PLANT AID, KGU	
	ANNUAL AID	CUMULATIVE AID
1985	3,785	64,403
1986	1,189	65,592
1987	(1,354)	64,238
1988	(299)	63,939
1989	(181)	63,758
1990	649	67,588
1991	649	68,237
1992	649	68,886
1993	649	69,535
1994	649	70,184
1995	649	70,833
1996	649	71,482
1997	649	72,131
1998	649	72,780
1999	649	73,429
2000	649	74,078
2001	649	74,727
2002	649	75,376
2003	649	76,025
2004	649	76,674
2005	649	77,323

FIGURE 3. URANIUM INVENTORY DIFFERENCE - PORTSMOUTH CUMULATIVE TOTALS, 1955 - 2005



modified in the late 1950s; this set of factors is referred to in the discussion below as the "old RI factors" shown in Figure 4.

The earlier calculational model made no allowance for changes in cascade operating conditions (such as temperature and pressure), factors that can strongly influence the rate of deposition of reduced uranium fluorides in the cascade. Consequently, a theoretical calculational model for cascade RI was developed in the early 1980s. Over the years, many laboratory corrosion studies have resulted in the development of equations describing the rates of reaction of UF_6 with nickel, aluminum, copper, steel and nickel-plated steel. Using these equations and the known surface areas of each cascade component, a calculational model to estimate the rate of formation of solid reduced uranium fluoride deposits (such as UF_5) in the cascade was developed. This model was sensitive to changes in cascade temperatures and pressures. In addition, the UF_6 adsorption on cascade surfaces, the chemisorption portion of which remains on the equipment surfaces when equipment is removed from service, is calculated by the model. This model, referred to as "new RI factors" in Figure 4, is believed to provide the best available estimate of the rate of RI accumulation in the present, post-CIP diffusion cascade. Note that the model makes no attempt to estimate the rate of deposition of uranium oxyfluorides formed by hydrolysis of UF_6.

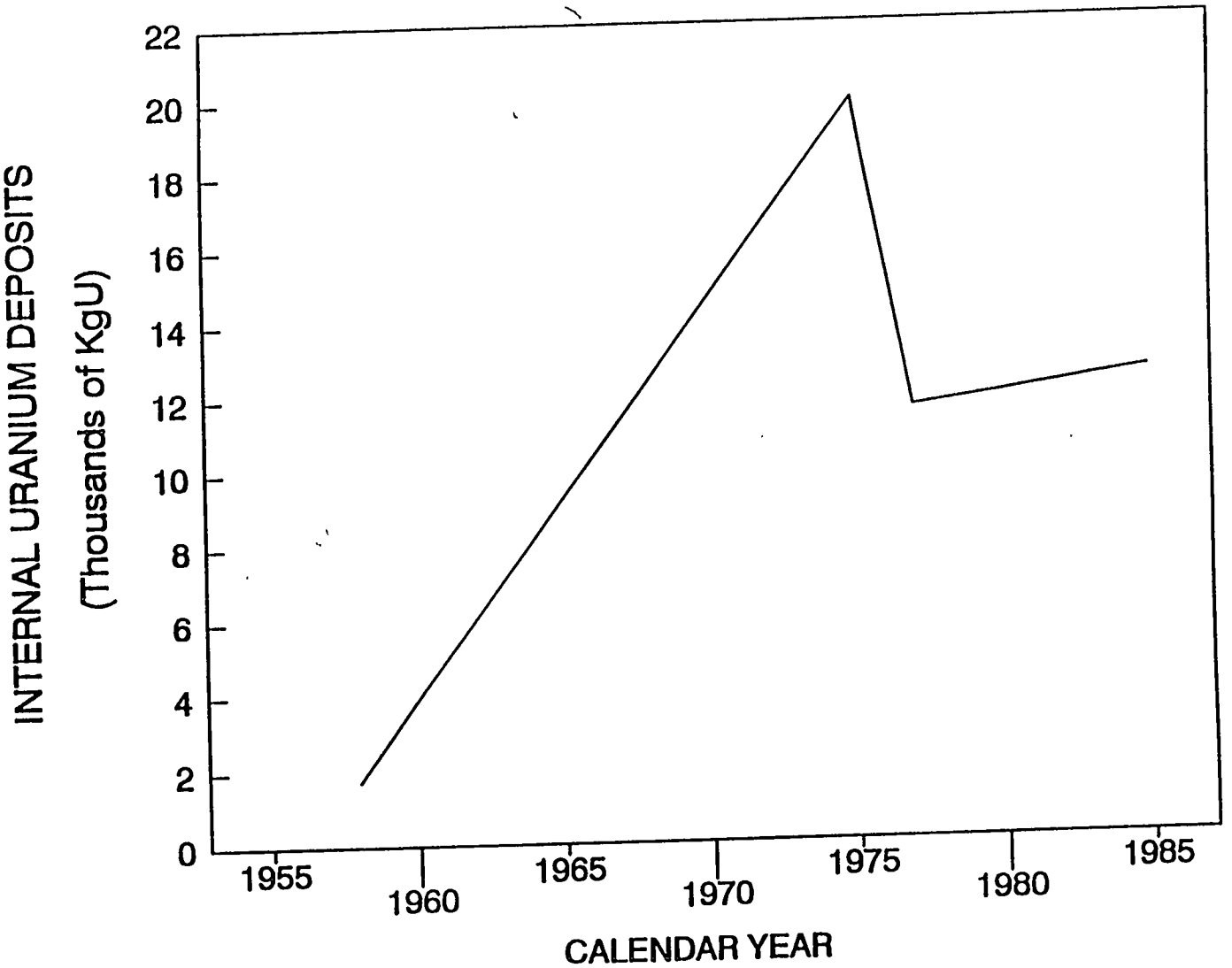
This new model predicts that most of the cascade surfaces will be maintained free of reduced uranium deposits such as UF_5. The reduced uranium fluoride deposits that are formed occur exclusively on bare steel surfaces. Many of these areas are located in recesses and crevices out of the main gas stream, which further aggravates the problem. A factor was built into the RI model to account for areas of nickel-plated steel in which the nickel plate may have failed for some reason. The calculated cumulative RI profile for the Oak Ridge cascade is shown in Figure 4. The values before 1975 were calculated using the "old RI factors" described earlier, while the new RI model was employed to calculate the values in the years after 1975. The decrease in the cascade ID of 8,500 Kg U observed during 1975 (as discussed earlier and as shown in Figure 1), was incorporated into the RI data of Figure 4 during the 1975-1977 time frame. The high rate of increases of RI before 1975 and the much lower rates calculated after 1977 are in good agreement with the ID data shown in Figure 1.

Based on the preceding discussion, the following conclusions covering the historical development of internal contamination in the gaseous diffusion plants can be made:

- As a result of immediate chemisorption of UF_6 on the cascade equipment surfaces, the need for eventual decontamination of equipment in gaseous diffusion plants was established when equipment was first exposed to UF_6.
- The equipment currently installed in the diffusion cascades exhibits a uranium deposition rate much lower than that exhibited by equipment operated in the 1950s and 1960s.
- This reduction in deposition rate was reflected by the much lower ID accumulation rate observed in the Oak Ridge cascade during its latter period of operation. Paducah and Portsmouth data show similar trends.
- Most uranium deposits in the current diffusion cascade occur either on areas of bare steel or in crevices and recesses that are not in the direct gas stream.

FIGURE 4. TOTAL INTERNAL URANIUM DEPOSITS - OAK RIDGE

OLD RI FACTORS THROUGH 1975, NEW RI FACTORS AFTER 1975



- At least a portion of the material chemisorbed upon initial exposure was on steel underlying the protective nickel plate, since the electroplated nickel plate exhibits significant porosity. To fully decontaminate the equipment, stripping off the nickel plate, which will be an expensive operation, will probably be necessary. The presence of additional uranium deposits is not expected to alter this cost significantly.

- The work required to D&D the cascades has not changed significantly since operation of the equipment began in the 1950s and 1960s. The levels of contamination within process equipment are considered to be rising at a very slow rate. In addition, the volumes of contaminated equipment have remained relatively unchanged.

NONDESTRUCTIVE ASSAY (NDA) SURVEYS OF PROCESS EQUIPMENT

Nondestructive assay (NDA) surveys are currently being conducted in the shutdown production facilities at The K-25 Site to locate residual deposits that may present a criticality concern during the D&D phase. This task is being performed by the Safeguards Studies Department of the International Technology Programs Division at Oak Ridge. The measurement survey consists of three major components: (1) isotopic mapping of selected items to identify the radionuclides present and to determine the isotopic composition of those nuclides, (2) scanning of the entire building to locate sizable deposits of uranium with in-process equipment and piping, and (3) quantitative measurement of large deposits.

Isotopic mapping involves conducting measurements using high-resolution gamma ray measurement instrumentation and then analyzing the spectra (1) to determine the ^{235}U concentration of uranium contained in process equipment and (2) to detect if unexpected isotopes that can interfere with NDA quantitative measurements are present. Deposits that can present a criticality concern are initially identified by scanning with gamma-ray measurement techniques. Neutron measurement methods were used to perform quantitative measurements of deposits identified during scanning operations.

Surveys have been completed in K-25 and K-27, the high-assay buildings, and are now being extended to the low-enriched uranium buildings. K-29 building was the first of the low-assay buildings to be surveyed, followed by K-31 and K-33. In keeping with the original intent, these surveys have been conducted to locate large deposits that might present criticality problems and not to totally characterize the internal contamination of process equipment.

The K-29 process building contains 300 diffusion stages, located in three units (sub-buildings) of 100 stages each. The equipment in these stages, converters and compressors, was upgraded during the initial improvement program in 1957-1962, but none was replaced during the CIP/CUP program of 1975-1982. Units 1 and 2 continued to operate until 1985, but Unit 3 was shut down in the spring of 1969 or about five years after K-25 and K-27 were shut down.

The uranium deposits identified in Building K-29 during the recent preliminary NDA survey are summarized in Table 4. Thirteen deposits averaging over 30 kgU each were found in the converters in Units 1 and 2, but only three deposits averaging 7 kgU were identified in Unit 3 converters. Another 280 kgU were found in 24 deposits within the compressors for an average size of less than 12 KgU per deposit. The interstage and intercell piping and booster stations found 3,600 KgU in 102 deposits for an average size of about 35 kgU per deposit. The K-29 converters were left untouched

TABLE 4. LOCATION OF SIZABLE DEPOSITS IN K–29*

UNIT	CONVERTERS/ COOLERS		COMPRESSORS		PIPING/ BOOSTER		TOTAL	
	NO.OF ITEMS	KGU	NO.OF ITEMS	KGU	NO.OF ITEMS	KGU	NO.OF ITEMS	KGU
1	7	207.4	4	39.5	38	1,849.4	49	2,096
2	6	228.2	3	28.7	30	1,435.0	39	1,692
3	3	20.8	17	215.1	34	315.2	54	551
TOTAL	16	456.4	24	283.3	102	3,599.6	142	4,465.3

* Process equipment was not replaced during CIP program

during the CIP program; so they have been onstream since 1960 or so. Unit 3 of K-29 was shut down in April 1969, some 16 years ahead of the remainder of the cascade that was shut down in 1985.

Uranium deposits identified in Building K-31 during the recent preliminary NDA survey are summarized in Table 5. As indicated in this table, only about half of the converters in this building were replaced during the CIP program and all of the others have been onstream since around 1960. Interstage and intercell piping have never been replaced. Over 95% of the deposits were located in the converters and piping. Less than 1% were located in compressors with only 4 of the 600 compressors containing deposits in excess of the threshold value of 1 kgU. Of the 2,491 kgU found in the converters, 2,332 kgU (94%) were in "old" converters while only 160 kgU (6%) were in the CIP converters. Thus, by far the majority of uranium deposits in converters were found in the "old" or pre-CIP equipment. Since the piping was not changed out during CIP, all of the deposits found there can be attributed to "old" equipment. Adding the 1,635 kgU found in the piping to 2,332 kgU found in the "old" converters, a total of 3,967 kgU can be attributed to "old" equipment. This represents 95% of the total deposits.

NDA survey of the K-33 building is currently under way. A change in survey procedure to undertake additional measurements in each cell has been made for K-33. Results for K-33 should be available later this year.

FEEDING OF REPROCESSED URANIUM

Feed material to produce UF_6 gas, which is the form used in the gaseous diffusion process, is derived mostly from natural uranium that has a ^{235}U content of 0.711%. For a period between 1953 and 1976, uranium used to fuel production reactors at Hanford and Savannah River was reprocessed to recover plutonium and uranium. This irradiated uranium at a slightly reduced enrichment was recycled by converting it into UF_6 and enriching it in the gaseous diffusion plants. A total of 101,268 metric tons of this reprocessed uranium was fed to the Paducah Gaseous Diffusion Plant[8] in combination with feed produced from natural uranium. Most of the recycled material (96%) had been irradiated in Hanford reactors. As shown in Table 6, reprocessed uranium represented over 13% of all material fed to the Paducah plant during this time, although in one year, 1973, it comprised 64% of the feed. In most years, however, reprocessed uranium constituted 25% or less of the total feed. This is also shown in Figure 5.

Over the same period, some 5,600 metric tons of reprocessed uranium from government production reactors were fed to the Oak Ridge Gaseous Diffusion Plant. Also, a much smaller quantity, 372 metric tons (or 4% of total reprocessed uranium), of much cleaner reprocessed uranium from commercial power reactors (reprocessed in France) was fed to the Oak Ridge GDP[9].

In addition to the minor uranium isotopes, ^{232}U, ^{234}U and ^{236}U, reprocessed uranium contains trace amounts of transuranic (TRU) elements such as neptunium and plutonium, and fission products such

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8. R. L. Ritter, et. al., *Neptunium Experience at PGDP*, K/ETO-30 (September 1990)

9. U. S. Department of Energy, *Options Analysis for Enrichment of Reprocessed Uranium*, K/ETM-711 (October 1985)

TABLE 5. LOCATION OF SIZABLE DEPOSITS IN K-31

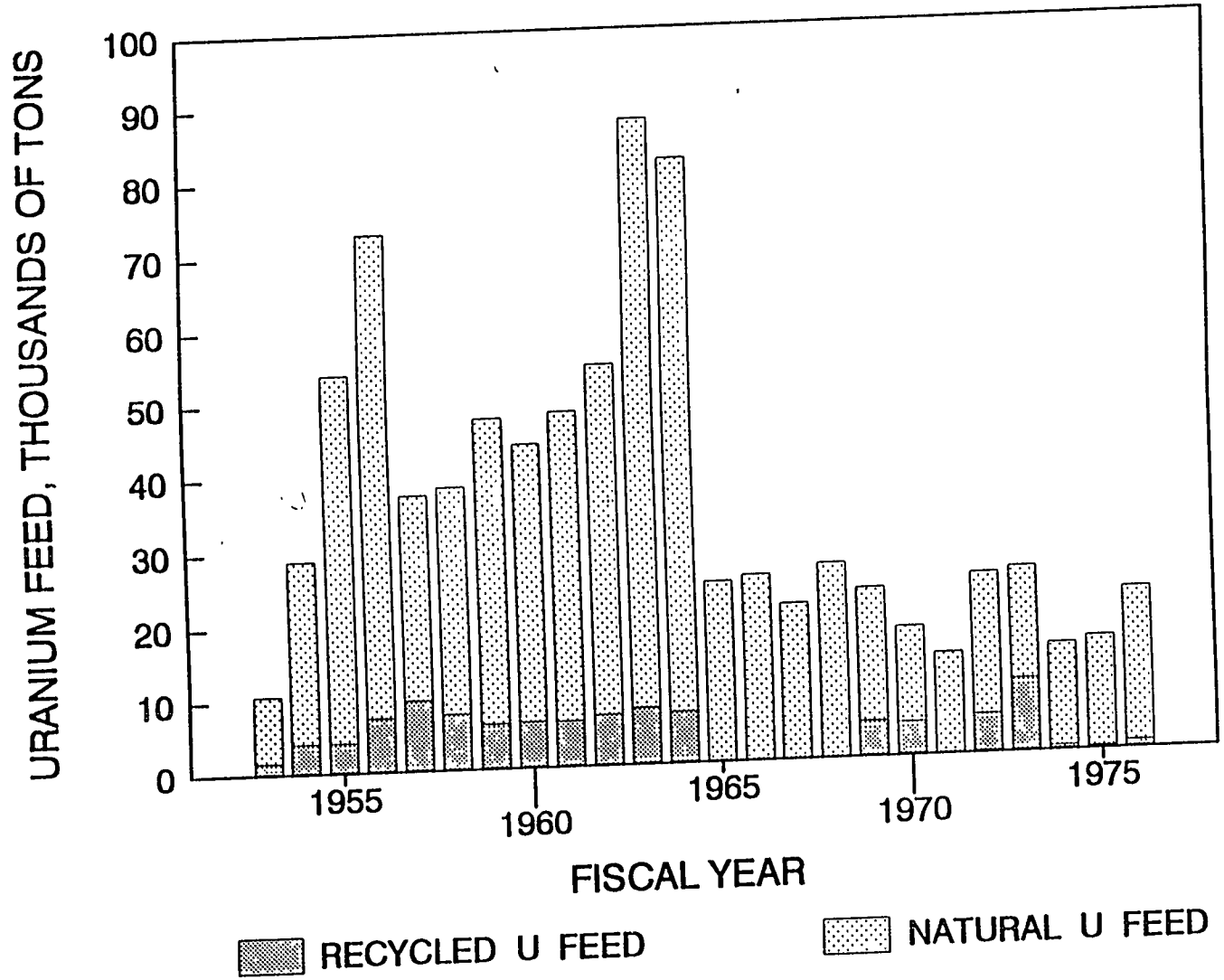
UNIT	CONVERTERS/ COOLERS		COMPRESSORS		PIPING/ BOOSTER		TOTAL	
	NO.OF ITEMS	KGU	NO.OF ITEMS	KGU	NO.OF ITEMS	KGU	NO.OF ITEMS	KGU
K-31 EQUIPMENT NOT REPLACED DURING CIP/CUP								
1	13	1,350.0	1	3.3	5	703.5	19	2,056.8
2 -1,3	3	150.9	0	0	4	275.3	7	426.2
3	0	0	0	0	3	79.0	0	0.0
4	0	0	0	0	2	174.4	0	0.0
5 even	24	466.9	0	0	5	167.3	29	634.2
6	12	363.7	0	0	6	235.7	18	599.4
TOTAL	52	2,331.5	1	3.3	25	1,635.2	73	3,716.6
K-31 EQUIPMENT REPLACED DURING CIP/CUP								
2-others	0	0.0	0	0	0	0	0	0.0
3	2	14.4	3	29.7	0	0	5	44.1
4	3	101.5	0	0.0	0	0	3	101.5
5 odd	2	43.8	0	0.0	0	0	2	43.8
TOTAL	7	159.7	3	29.7	0	0	10	189.4

TABLE 6. REPROCESSED URANIUM (RU) FEED AT PADUCAH

FY	RU TONS U	NATURAL U FEED	TOTAL FEED TONS U	RU % TOTAL
1953	1,565	7,587	9,152	17.1%
1954	4,104	20,675	24,779	16.6%
1955	4,066	45,769	49,835	8.2%
1956	7,383	58,101	65,484	11.3%
1957	9,674	17,995	27,669	35.0%
1958	7,653	23,029	30,682	24.9%
1959	6,193	34,993	41,186	15.0%
1960	6,317	31,138	37,455	16.9%
1961	6,217	35,642	41,859	14.9%
1962	6,978	40,392	47,370	14.7%
1963	7,745	71,965	79,710	9.7%
1964	7,003	67,997	75,000	9.3%
1965	0	24,611	24,611	0.0%
1966	0	25,334	25,334	0.0%
1967	0	21,251	21,251	0.0%
1968	0	26,567	26,567	0.0%
1969	4,781	13,474	18,255	26.2%
1970	4,529	8,543	13,072	34.6%
1971	0	13,895	13,895	0.0%
1972	5,283	13,990	19,273	27.4%
1973	9,904	5,402	15,306	64.7%
1974	500	13,693	14,193	3.5%
1975	415	14,608	15,023	2.8%
1976	958	20,083	21,041	4.6%
TOTAL	101,268	656,734	758,002	13.4%

FIGURE 5. RECYCLED URANIUM AND NATURAL URANIUM FEED

PADUCAH - 1953 to 1976



as ^{99}Tc. These radioactive isotopes not only present significant problems by degrading the purity of the final product but also pose a radiation hazard to the work force. While relatively clean reprocessed uranium can be prepared as was done in France, this was not the case with Hanford and Savannah River material. Technetium concentrations in reprocessed uranium from government production reactors were found to have between 175 and 700 times the levels found in the reprocessed uranium from commercial reactors. In addition, concentrations of transuranics in uranium from U.S. production reactors were 20 to 450 times greater than the concentrations in commercial reprocessed uranium. Thus, the production reactor's uranium contributed more than 99% of the technetium and transuranics fed to the gaseous diffusion plants.

Technetium is very close to uranium in its chemical nature and forms compounds that carry through the chemical separation processes to a significant degree[10]. A number of its compounds, such as TcF_6, are gaseous under cascade conditions and being of lower molecular weight than UF_6, diffuse preferentially toward the enriching sections of the cascade. However, this transport of technetium was found to take place rather slowly; technetium compounds are deposited on internal surfaces of the cascade equipment and then, over a period of several years, migrate toward the enriching portion. In the case of Paducah, technetium was found in the Paducah product about two years after the feeding of reactor return materials started.

Some of the technetium initially fed at the Paducah plant was later shipped on to Portsmouth and Oak Ridge as part of the Paducah product feed to those diffusion plants. As a result, contamination at all three diffusion plants, not only with uranium but also with technetium and transuranics, can be ascribed to the government's operating policy during the early years. While only a fraction of the transuranics and technetium present in the reprocessed uranium actually enters the cascade and some such as Pu remain relatively fixed near the feed point to the cascade, others tend to migrate throughout all the plants.

A significant portion of these radioactive isotopes is considered to have been extracted from the cascade by the two cascade improvement programs, one in the sixties and another in the late seventies when a major portion of the barrier tubes was replaced under both programs with higher performance tubes. However, traces of these radioactive isotopes are still to be found today in cascade equipment.

TRU limits based on DOE Order 5480.11 are 20 dpm and 300 dpm for transferrable and total contamination, respectively. These lower limits could cause more material to be declared as contaminated as opposed to the uranium limits of 1,000 dpm and 5,000 dpm alone. Some structures, materials and equipment that have not been classified as contaminated may have to be declared contaminated at the relatively higher criteria levels for uranium.

The transuranics contaminates are a major determinant in radiation protection practices at Paducah and are expected to complicate the final D&D process because transuranics are more radiotoxic, and thus controls are more restrictive for D&D activities. The use of respirator and protective clothing and equipment would have to be greater and would impact productivity of the workers carrying out D&D activities and also would increase the cost of labor and supplies. Analytical costs for TRU are

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10. J. C. Bailey, *Radionuclides in the Equipment of the Oak Ridge Diffusion Plant*, March 10, 1988 (Draft)

greater than those for uranium, and quality control requirements are more extensive. Bioassay for TRU is more difficult to perform and is more expensive than for uranium. Worker training in radiation protection for TRU and health physics support are more extensive and would cost more for D&D activities. Management options are more limited for TRU contaminated waste and storage and disposal may be required at increased cost at a site specifically designed for transuranics.

Thus, contamination of all of the gaseous diffusion plants by these transuranic elements and fission products was clearly the direct result of feeding reprocessed uranium from the U. S. government's defense production reactors into the gaseous diffusion plants.

CONTAMINATION OF BUILDING INTERIORS

EQUIPMENT EXCHANGES

Gaseous diffusion plants perform the uranium enrichment process by passing the process gas in the form of UF_6 through thousands of stages connected in series, with a small isotopic enrichment occurring in each stage. Each stage consists of a compressor that forces process gas through a pressure vessel called the converter and on to the next stage. The converter contains thousands of porous tubes that constitute the separating membrane. The converter also contains a heat exchanger, the gas cooler, that removes the heat of compression created by the compressor and maintains the process at the optimum operating temperature. Other major components are electric motors that drive compressors and interstage piping and valves that direct the flow of the process gas from between stages.

The low-assay portion of gaseous diffusion cascades perform the task of enriching natural uranium from its initial assay of 0.711% ^{235}U to assays ranging from 3% to 5% for use as fuel in commercial nuclear power reactors. The reject or depleted stream exits the bottom of the cascade at an enrichment ranging from 0.20-0.40%. Three different sizes of process equipment are employed in the low-assay portion of the cascades. These are commonly referred to as 0, 00 and 000 size equipment with the 000 being the largest size. These sections of the cascades are often referred to as axial stages or axial equipment because the compressors used are of the axial flow type as differentiated from the centrifugal flow compressors employed in the high-assay section of the plants.

The cascade at Oak Ridge, which has been out of operation since 1985, consists of three buildings: K-29, which contains 300 stages of the 0 size equipment, K-31 with 600 stages of the 00 size and K-33 with 640 stages of 000 size equipment. The 300 stages in K-29 are located in three sub-buildings called units of 100 stages, each of which is further subdivided into ten cells of ten stages each. The cell is the smallest subdivision of operating equipment that can be operated by itself or be bypassed (taken off-stream) and shut down without interrupting the flow of process gas through the remainder of the cascade. The K-31 building is subdivided into six units of ten cells each with ten stages in each cell. The 640 stages in K-33 are subdivided into eight units of ten cells each, with eight stages per cell. Thus, 1,540 diffusion stages are located in the three low-assay process buildings at Oak Ridge.

The low-assay cascade at Portsmouth is very similar to Oak Ridge in size and arrangement. The X-29 building consists of six units, each containing ten cells of ten stages each, a total of 600 stages of 0

size equipment. The X-31 building is subdivided into five units of ten cells each for a total of 500 stages of 00 size equipment. At Portsmouth, the X-29 and X-31 buildings are combined into a single large facility, the X-330 building. The 000 size equipment at Portsmouth is located in the X-333 building, which is very similar to the K-33 building in that it has 640 stages subdivided into eight units of ten cells each, with eight stages per cell. The Portsmouth low-assay cascade totals 1,740 stages.

The Paducah cascade consists of four buildings: C-331, C-333, C-335 and C-337. C-331 and C-335 are alike in that they each contain 400 stages of 00 size equipment. The 400 stages are subdivided into four units within each of the buildings with ten cells per unit and ten stages per cell as in the -31 buildings at Oak Ridge and Portsmouth. C-333 and C-337 buildings are alike in that each contains 480 stages of 000 size equipment, with each building subdivided into six units of ten cells, eight stages per cell. Paducah has a total of 1,760 diffusion stages of 00 and 000 sizes.

Most gaseous diffusion equipment is operated at subatmospheric pressure so that outleakage of slightly radioactive process gas is not a major problem. Outleakage does occur on occasion, however, particularly when there is some type of misoperation or rupture of a connecting tubing during a transfer operation such as for the feed, product, or tails handling operations. Most of the process pipe connections are welded to minimize the possibility of leaks either into or out of the process. A patented seal design is used at the compressor where the drive shaft from the motor penetrates the compressor shell to connect with the rotor.

With the large number of equipment and the 35 to 45 years of operating history, inevitably equipment has been removed on numerous occasions from process buildings to decontamination and maintenance buildings for repairs, replacement or upgrading. Each time that a piece of stage equipment must be repaired or replaced, the smallest operating unit, the eight- or ten-stage cell, must be shut down, evacuated to remove process gas from within the cell, and refilled with dry air or dry nitrogen to provide an inert atmosphere during the removal process. In spite of these precautions, however, some particles of uranium that have reacted with moist air to form UO_2F_2 are typically released in the area where process piping has been cut to remove the chosen equipment from the cell. In the earlier days of operation, small plumes of smoke were often seen as remaining trace amounts of process gas reacted with moisture in the air to form particles of UO_2F_2 that settled in the vicinity of the removal operation. The amount and degree of contamination spread are dependent on several factors. Improvements have been made over time to reduce the potential for radioactive material escaping when equipment is opened for repair. The extent of contamination from such releases depends upon the following factors:

- Air current created by high volume building ventilation fans can disperse the material
- A small release will tend to deposit uranium on the surface of equipment in the vicinity of the leak
- A large release may well be swept by air currents to other parts of the building.
- A release at locations above cascade feed points will tend to contain traces of other radioactive materials such as technetium.
- Deposits may be transferrable contamination at first, but with time may penetrate the area of deposition and become fixed within the matrix media.
- Releases from some equipment may leach into the area over which the spill occurred and may become more difficult to decontaminate by conventional cleaning methods.

In addition, when radioactive material escapes its containment and is deposited on building structures, the possibility exists that it will not be discovered and/or completely removed by cleaning. In more recent periods, however, improvements in procedures have been made to prevent radioactive materials from being released, or if such releases occur, greater attention is given to cleaning such deposits and spills.

While alpha-emitting uranium is not considered particularly hazardous, it is recognized as being harmful if a person were to ingest it, primarily by breathing contaminated air. Thus, an extensive bioassay program of urinalysis has been maintained throughout the operating history to ensure that individual exposures are limited. Most recently, extensive upgrades in contamination control and radiation practices have significantly reduced even small exposures.

As described, opportunities for contaminating some of the areas in process buildings were many with the frequent transfer of process equipment in and out of process buildings. To this end, a review of movements of process equipment in and out of cascade buildings was made to ascertain the number of occasions for such transfers. In addition to a large number of equipment failures, especially in the early years, two major improvement programs of process equipment resulted in a large number of equipment movements. The process of removing this equipment opened the otherwise enclosed cascade to the atmosphere and created the potential for release of process gas and deposits of uranium-bearing compounds. The transport of equipment to various decontamination and maintenance facilities allowed further opportunities for spread of contamination. Upgrade and repair work on the equipment provided further opportunities for contamination of facilities.

The original plants at Oak Ridge, Paducah, and Portsmouth were built and placed into operation in the late forties and early to mid-fifties. Starting about 1956, an initial cascade Improvement Program was undertaken because significant improvements in the separating membrane had been made over those initially installed. At the same time, improvements in compressor technology had also been achieved so that in the period from 1956 to 1962, essentially all of the equipment, both converters and compressors, in the low-assay portion of the cascade was replaced with higher-performance equipment. This program was termed the Improvement Program. As laboratory work continued, still more efficient membranes were developed that justified another equipment changeout program termed the CIP/CUP program, which occurred from 1975 to 1982. Thus, there were two very large equipment changeout programs in the low-assay diffusion cascades in addition to the many exchanges of failed equipment over the course of the years.

Table 7 is a listing of the number of compressor, converter and seal exchanges that occurred at Oak Ridge between 1951 and 1985[11,12,13]. All three sizes, 0, 00, and 000, have been combined into this table to indicate the number of exchanges that have occurred over the period of operation of the Oak Ridge cascade. The list has been broken into pre-1969 equipment exchanges and post-1969 equipment exchanges as a differentiation between the defense era and the combined defense and

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11. D. C. Lannum, Informal communication, "Three Site Equipment Failure for Compressors, Seals, Converters", October 24, 1991

12. *Process Equipment Modification Program Completion Report*, May 1985 (K/BD-1071)

13. *ORGDP Plant Quarterly Reports*, FY 1955 through FY 1966

TABLE 7. AXIAL EQUIPMENT FAILURES AND REPLACEMENTS AT OAK RIDGE

FY	Axial Compresor Fail.&Repl.	Converter Fail.&Repl.	Seal Fail.&Repl.	TOTAL
1951	50	0	3,734	3,784
1952	342	0	3,996	4,338
1953	501	0	2,837	3,338
1954	210	0	6,444	6,654
1955	370	166	5,880	6,416
1956	238	590	2,948	3,776
1957	291	302	2,492	3,085
1958	552	320	2,324	3,196
1959	303	104	1,516	1,923
1960	39	135	1,219	1,393
1961	48	27	307	382
1962	32	29	111	172
1963	42	56	161	259
1964	89	24	323	436
1965	30	32	130	192
1966	66	8	187	261
1967	24	0	143	167
1968	47	0	151	198
1969	29	0	143	172
SUBTOTAL	3,303	1,793	35,046	40,142
1970	47	0	263	310
1971	137	0	377	514
1972	122	0	485	607
1973	44	0	321	365
1974	37	0	326	363
1975	27	20	432	479
1976	204	181	808	1,193
1977	317	283	1,153	1,753
1978	324	154	1,048	1,526
1979	280	188	937	1,405

TABLE 7. AXIAL EQUIPMENT FAILURES AND REPLACEMENTS AT OAK RIDGE (cont.)

FY	Axial Compresor Fail.&Repl.	Converter Fail.&Repl.	Seal Fail.&Repl.	TOTAL
1980	253	203	859	1,315
1981	216	150	702	1,068
1982	3	2	151	156
1983	6	3	122	131
1984	0	1	112	113
1985	4	2	115	121
1986	0	3	0	3
SUBTOTAL	2,021	1,190	8,211	11,422
GRAND TOTAL	5,324	2,983	43,257	51,564

commercial production era. Seal failures were included in this table because most seal failures, while correctable in place in most cases, can lead to local contamination resulting from escaping gas or particles falling to the floor when a seal exchange is performed. As can be seen in this table, the occasions for contaminating areas of process buildings were numerous. Some 40,100 equipment replacements occurred before 1970 and another 11,400 after 1970.

Figure 6 is a graphical representation of the number of compressor and converter exchanges that were performed and that were caused by failures and by upgrading to an improved design for Oak Ridge. It depicts the large replacement activity at Oak Ridge in the mid-to-late fifties. A somewhat lower level of compressor exchanges, some 2,000 including the CIP/CUP program, took place after 1970 as compared to 3,300 exchanges between 1951 and 1969. Almost 1,800 converter exchanges were performed between 1951 and 1970 compared to 1,190 between 1970 and 1985. Under each program, contamination of areas of process buildings is known to have occurred, although measurements of such contamination were not made or systematically recorded. Figure 7 shows the number of seal exchanges that occurred in the Oak Ridge low-assay cascade. Because of the very large number of seal failures experienced in comparison to numbers of compressor and converter failures (43,000 versus 8,300), seal failures are presented in a separate plot. The large number of seal failures during the very early years of operation led to a very high number of 35,000 seal replacements before 1969 compared to 8,200 exchanges after 1970.

Table 8 shows the history of the number of compressor, converter and seal replacements performed at the Paducah plant between 1954 and 1990[11,12,14]. A total of 15,600 exchanges were performed before 1970 as compared to 14,800 from 1970 to 1990. Figure 8 shows the number of compressor and converter exchanges at Paducah over the years; 2,070 compressor exchanges took place on or before 1969 versus 2,160 in the years between 1970 and 1990. The number of converter replacements was 3,100 prior to 1970 and 1,600 in the years after. Figure 9 gives the number of seal replacements: 10,400 were performed from 1954 to 1969 and 11,000 from 1970 and 1990.

Table 9 provides the number of compressor, converter and seal exchanges that were performed at the Portsmouth plant low assay facilities between 1954 and 1990[11,12]. Some 35,800 replacements took place between 1954 and 1969, and another 13,000 were performed between 1970 and 1990. Figures 10 and 11 are graphical presentations of the number of compressor plus converter replacements and of seal replacements, respectively, performed at Portsmouth.

A tremendous number of equipment exchanges have been performed at the Oak Ridge, Paducah, and Portsmouth diffusion plants since the start of operation at each of the sites. Each exchange is considered to be at least an opportunity for the spread of uranium contamination around working areas on cell floors. Routine measurements of contaminates were not made or recorded systematically to document the level of area contamination existing before and after each of the exchanges. However, the occurrence of contamination is inescapable, and the start of uranium contamination in process buildings can be traced to the large number of equipment exchanges caused by failures that began in the very early years and that were augmented by the first Improvement Program for compressors and converters. This period, 1951 to 1964, was the time when uranium enrichment was performed solely for defense purposes. Because contamination of process buildings was caused by early efforts to replace process equipment, the fact that decontamination of process buildings and

[14] *Paducah Plant Quarterly*, FY 1955 through FY 1966 (KY-128 et.al.)

FIGURE 6. FAILURES AND REPLACEMENTS - OAK RIDGE AXIAL COMPRESSORS AND CONVERTERS

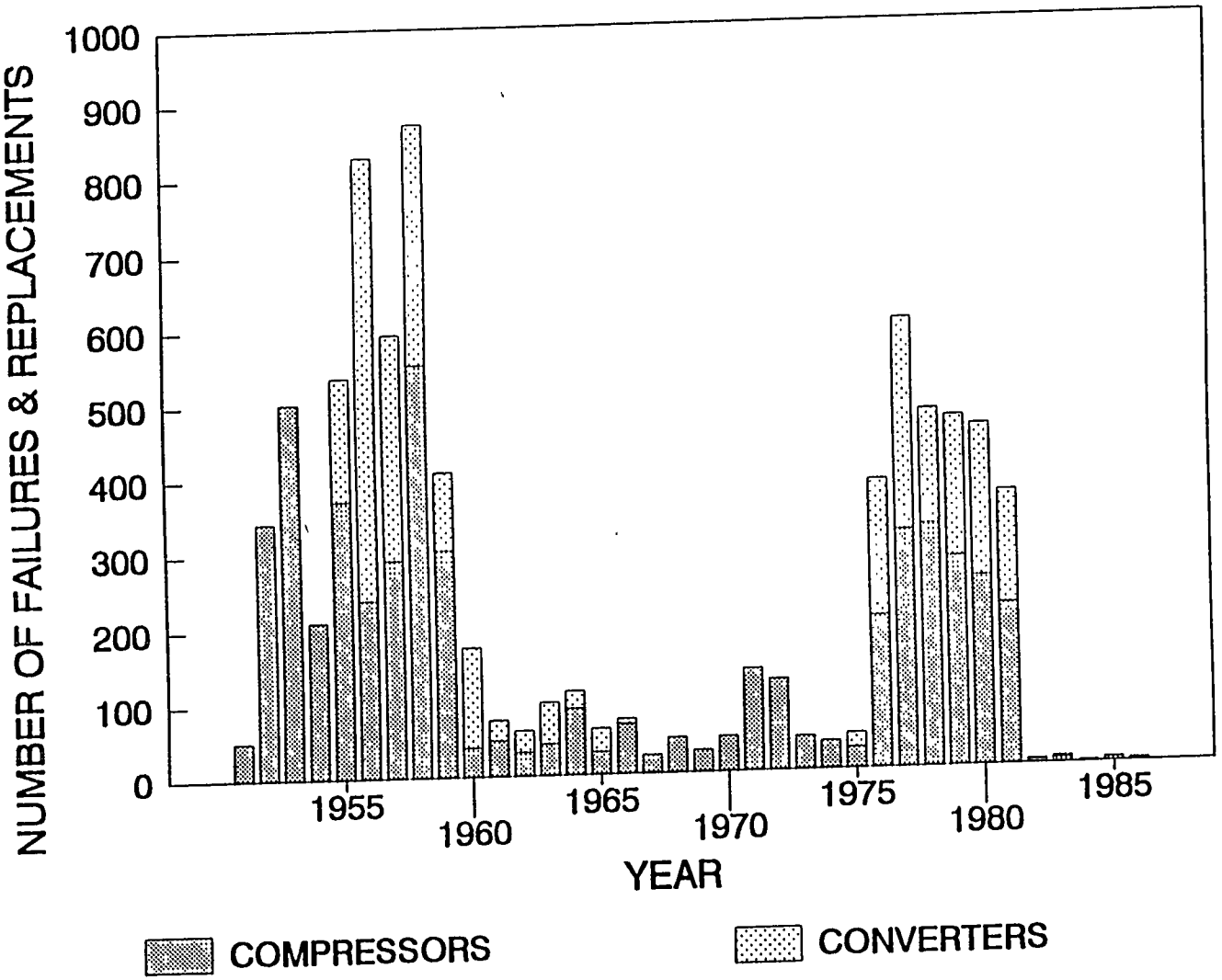


FIGURE 7. SEAL FAILURES & REPLACEMENTS- OAK RIDGE AXIAL COMPRESSOR SEALS

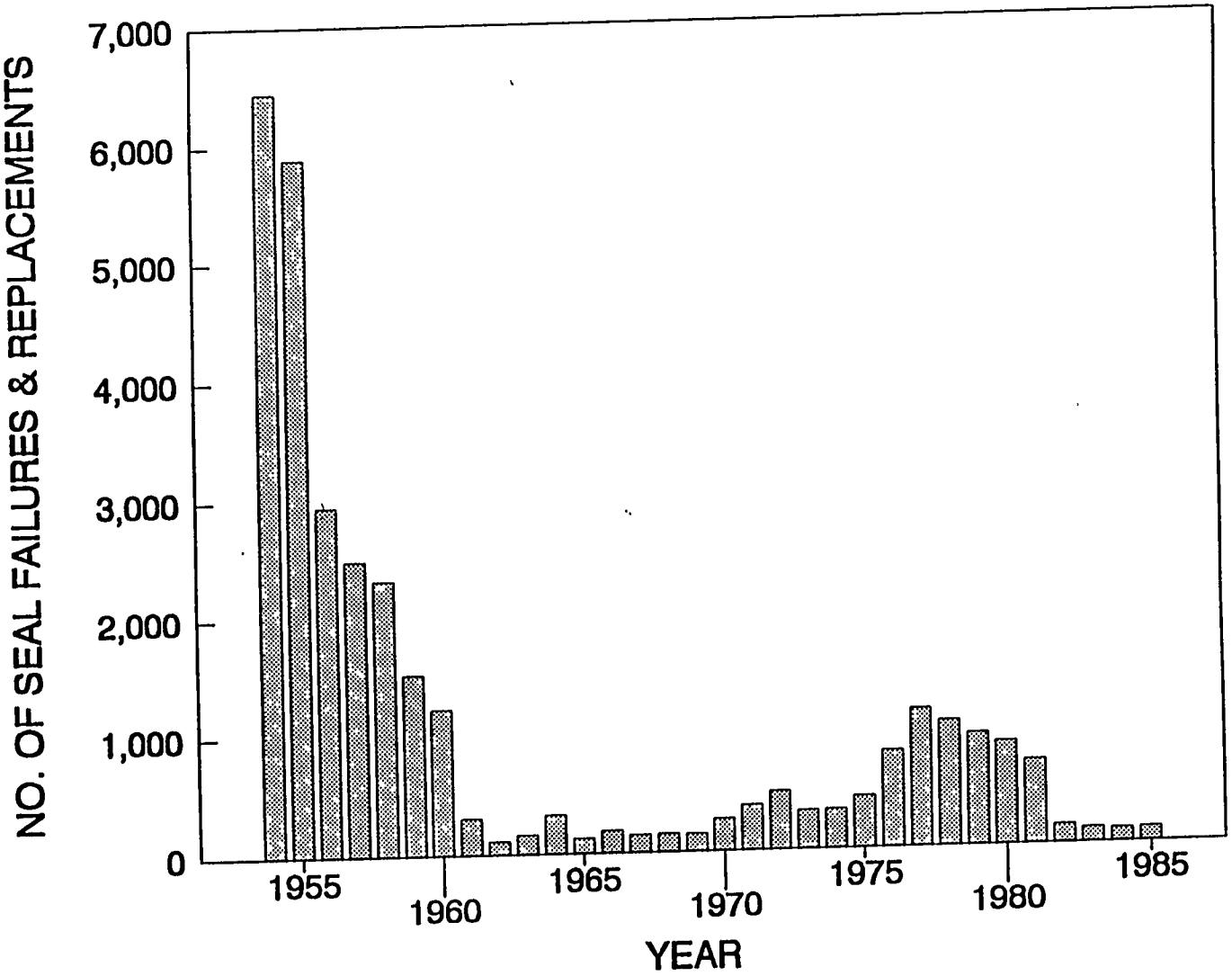


TABLE 8. AXIAL EQUIPMENT FAILURES AND REPLACEMENTS AT PADUCAH

FY	Axial Compresor Fail.&Repl.	Converter Fail.&Repl.	Seal Fail.&Repl.	TOTAL
1954	22	11	0	33
1955	313	389	2,356	3,058
1956	355	545	2,029	2,929
1957	379	520	1,295	2,194
1958	96	346	634	1,076
1959	276	344	727	1,347
1960	136	299	597	1,032
1961	54	301	490	845
1962	84	136	239	459
1963	75	80	317	472
1964	67	75	355	497
1965	28	11	314	353
1966	17	9	149	175
1967	61	21	365	447
1968	42	24	289	355
1969	63	12	242	317
SUBTOTAL	2,068	3,123	10,398	15,589
1970	54	26	348	428
1971	16	9	359	384
1972	18	10	321	349
1973	29	22	401	452
1974	19	17	203	239
1975	30	56	433	519
1976	292	254	1,072	1,618
1977	408	407	1,390	2,205
1978	418	339	1,469	2,226
1979	311	290	1,038	1,639
1980	333	200	1,071	1,604
1981	196	2	841	1,039
1982	4	0	455	459

TABLE 8. AXIAL EQUIPMENT FAILURES AND REPLACEMENTS AT PADUCAH (cont.)

FY	Axial Compresor Fail.&Repl.	Converter Fail.&Repl.	Seal Fail.&Repl.	TOTAL
1983	6	0	415	421
1984	0	0	296	296
1985	7	1	149	157
1986	8	2	107	117
1987	3	1	143	147
1988	3	0	131	134
1989	4	0	183	187
1990	6	0	221	227
SUBTOTAL	2,165	1,636	11,046	14,847
GRAND TOTAL	4,233	4,759	21,444	30,436

FIGURE 8. FAILURES AND REPLACEMENTS - PADUCAH AXIAL COMPRESSORS AND CONVERTERS

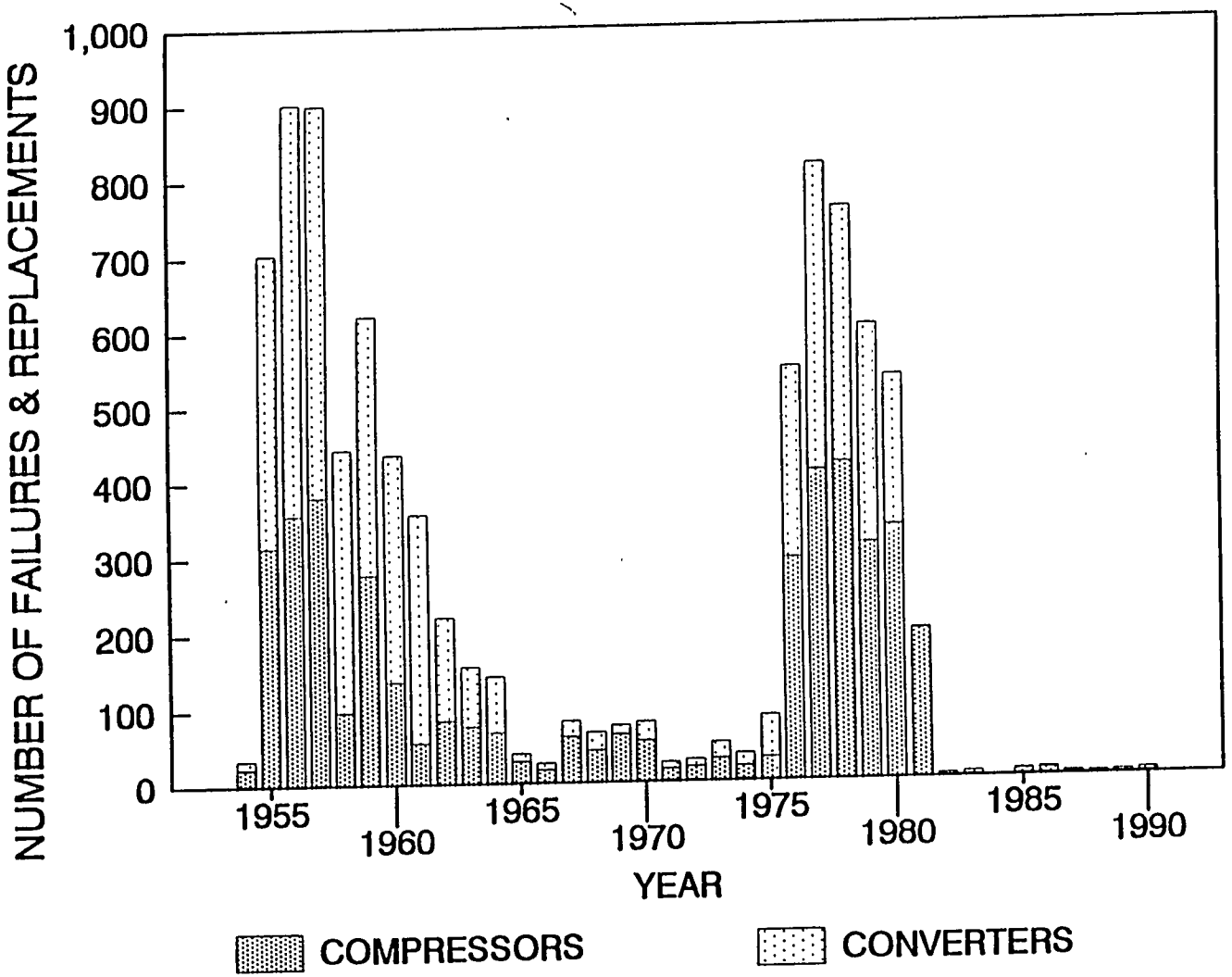


FIGURE 9. FAILURES AND REPLACEMENTS - PADUCAH AXIAL COMPRESSOR SEALS

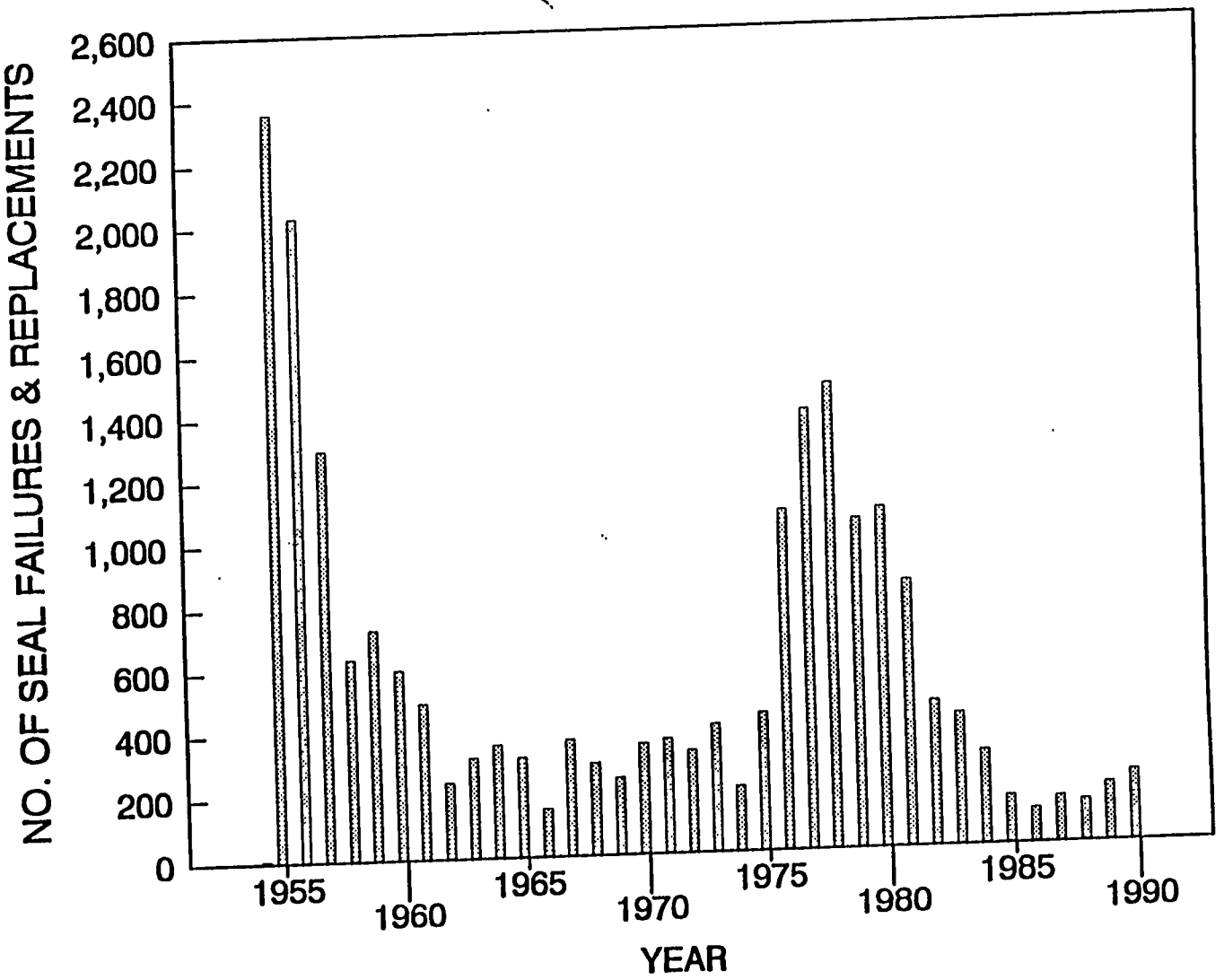


TABLE 9. AXIAL EQUIPMENT FAILURES AT PORTSMOUTH

FY/CY	Axial Compresor Failures	Converter Failures	Seal Failures	TOTAL
1954	64	10	2,574	2,648
1955	26	16	5,516	5,558
1956	59	77	4,700	4,836
1957	79	480	5,169	5,728
1958	196	374	5,402	5,972
1959	240	216	2,134	2,590
1960	498	294	1,563	2,355
1961	478	160	1,160	1,798
1962	273	61	595	929
1963	183	70	431	684
1964	147	61	478	686
1965	150	50	469	669
1966	74	24	372	470
1967	38	34	293	365
1968	22	18	217	257
1969	65	25	245	335
SUBTOTAL	2,592	1,970	31,318	35,880
1970	51	23	216	290
1971	42	21	352	415
1972	76	14	326	416
1973	49	38	240	327
1974	74	47	333	454
1975	216	124	521	861
1976	222	137	714	1,073
1977	344	203	892	1,439
1978	273	212	980	1,465
1979	134	105	481	720
1980	123	126	329	578
1981	236	270	620	1,126
1982	208	216	673	1,097
1983	91	30	317	438
1984	56	30	396	482

TABLE 9. AXIAL EQUIPMENT FAILURES AT PORTSMOUTH (cont.)

FY/CY	Axial Compresor Failures	Converter Failures	Seal Failures	TOTAL
1985	69	37	425	531
1986	29	50	243	322
1987	16	27	315	358
1988	6	0	105	111
1989	30	0	195	225
1990	37	0	298	335
SUBTOTAL	2,382	1,710	8,971	13,063
GRAND TOTAL	4,974	3,680	40,289	48,943

FIGURE 10. FAILURES & REPLACEMENTS - PORTSMOUTH AXIAL COMPRESSORS AND CONVERTERS

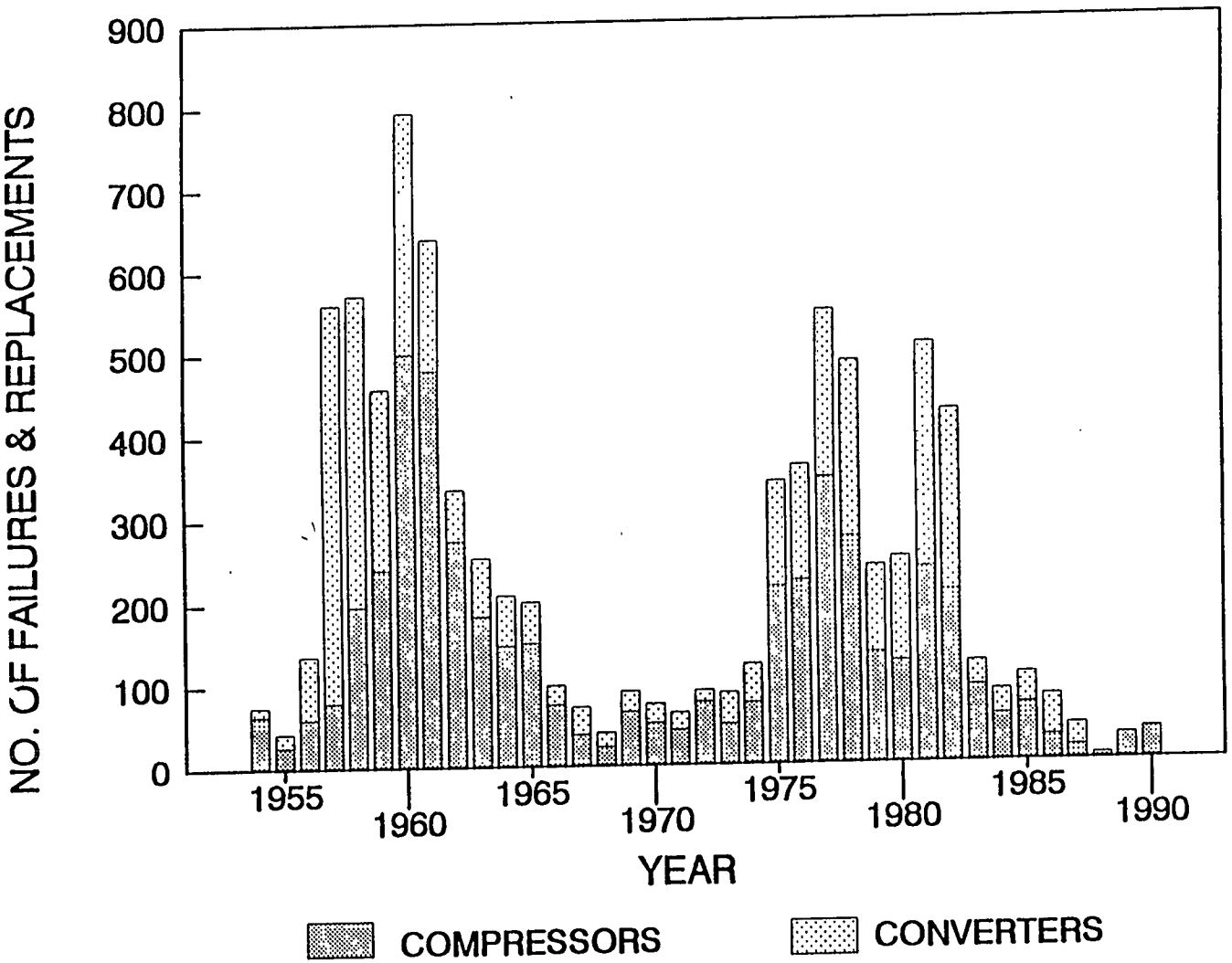
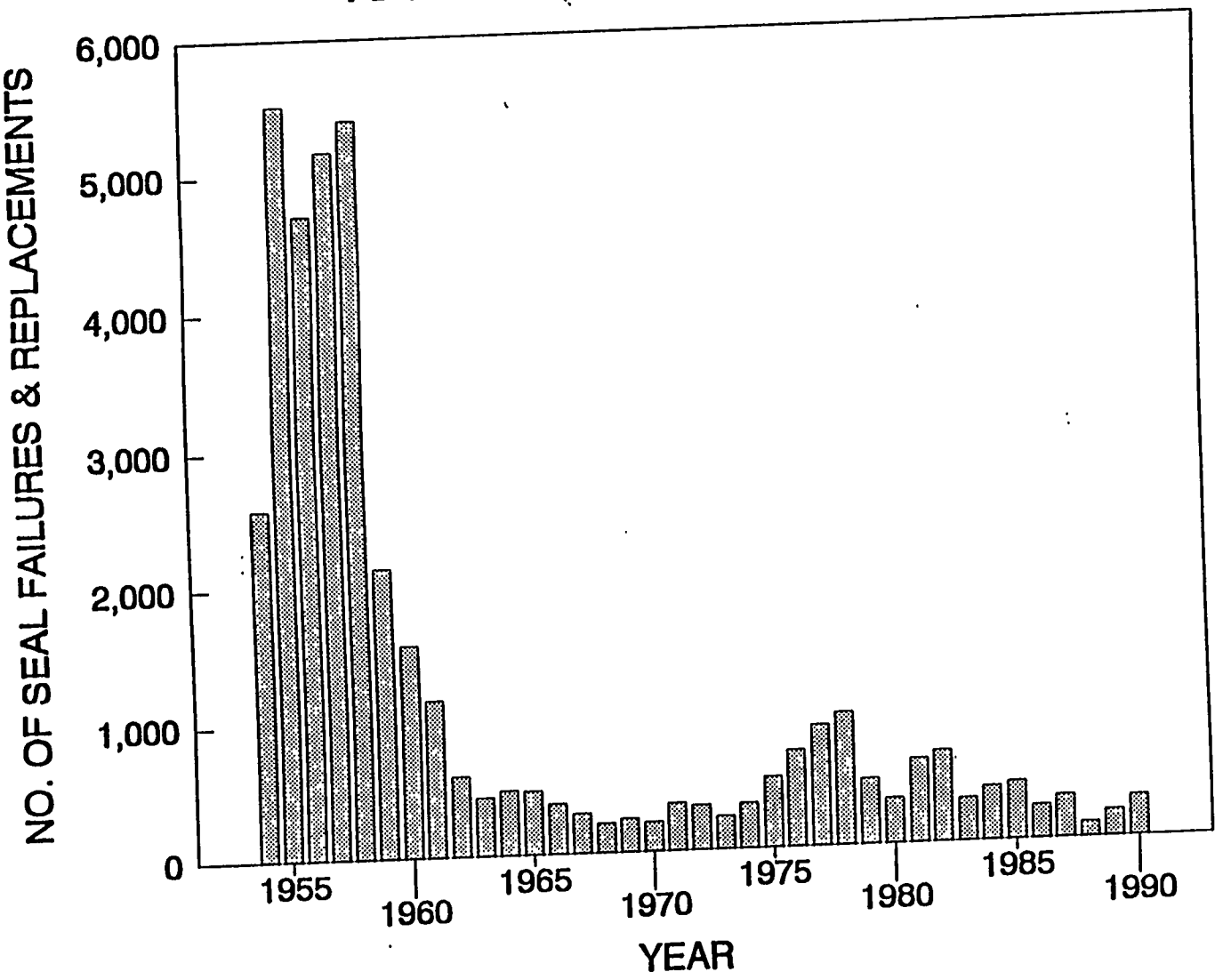


FIGURE 11. SEAL FAILURES & REPLACEMENTS-PORTSMOUTH AXIAL COMPRESSOR SEALS



most auxiliary buildings was required is also believed to have been established. Further exchanges of process equipment made only incremental contributions to existing contamination.

With today's contamination control practices and programs to clean past contaminated areas, the opportunity for further spread of contamination caused by equipment replacement has been essentially eliminated.

PROCESS GAS RELEASE INFORMATION

Since the start of operation of the gaseous diffusion plants, accidental releases of UF_6 gas have occurred from time to time as a result of a number of causes. These releases contributed to contamination within the various buildings where they occurred and to external surroundings. Records of these accidental releases that involved quantities greater than 1 kg have been provided by each of the three sites in terms of the date of occurrence, nature of the incident, and estimated amount of UF_6 released. Smaller gas releases were not recorded until relatively recently. When a release of process gas, UF_6 gas, occurs, it reacts almost instantaneously with moisture in the air to form UO_2F_2, a solid in the form of fine particles. These particles are deposited in the immediate area or are carried by air currents to surrounding areas. Thus, contamination with uranium can be confined to a relatively small area such as within a cell housing, or it can be widespread if the incident occurred in an open area inside a building and the particles are swept into the building ventilation system. An incident outside a building can release materials to the atmosphere, and the materials can be deposited nearby or carried by wind to other areas. Material deposited on building floors or outside can be tracked into nearby buildings on shoes of employees. In the early days of GDP operations, precautions against the spread of contamination were not implemented as rigorously as current rules require. Since the data described in this section indicate a much higher number of large releases occurring in the early days of GDP operation for government purposes, responsibility for contamination of and within buildings can clearly be assigned to the earlier government production operations.

Under the new contamination control program, operations and maintenance programs have adopted a zero-release goal that has dramatically reduced the number and size of process gas releases and subsequent spreading of that contamination. The historical data have been summarized for Oak Ridge, Paducah and Portsmouth diffusion plants for cascade buildings, feed plant and all other buildings but excluding high assay portions of the cascades (K-25, K-27 and X-326 buildings). Process gas releases occurring within the high-assay buildings are considered to be related only to the contamination levels within these buildings that are used solely for government production. Cross-contamination between buildings is considered to be highly unlikely and impossible to verify.

The information has been broken into releases occurring within process buildings, the feed facility, and all other facilities. The feed facility was tabulated separately because of the relatively large number as well as the significant sizes of releases occurring there. The many other facilities on the GDP sites contribute a sizeable portion of releases of process gas through the multitude of activities that involve the handling of UF_6 gas. These releases into the atmosphere are considered to have resulted in contamination within buildings as well as to outside the buildings when the UF_6 hydrolyzes to UO_2F_2 and is deposited in the immediate area.

The magnitude of releases at ORGDP for each year since 1951 is shown in Table 10 for process buildings (K-29, K-31 and K-33), for K-1131, the feed facility, and for all other buildings (except K-25

TABLE 10. SUMMARY OF PROCESS GAS RELEASES AT ORGDP*

YEAR	KG. OF PROCESS GAS RELEASED			
	K–29,K–31 K–33	K–1131	ALL OTHERS	TOTAL
1951	1.23	104.94	82.61	188.78
1952	0.00	322.05	852.43	1,174.48
1953	0.00	289.70	327.45	617.15
1954	0.00	0.00	42.41	42.41
1955	146.66	3.07	169.91	319.64
1956	2.74	30.67	186.52	219.93
1957	0.00	3.37	3.04	6.41
1958	26.63	1,173.89	30.20	1,230.72
1959	0.00	0.00	2.10	2.10
1960	0.00	858.98	43.35	902.33
1961	1.84	108.83	198.43	309.10
1962	0.00	0.00	26.01	26.01
1963	0.00	0.00	0.00	0.00
1964	0.00	3.07	3.07	6.14
1965	0.00	0.00	4.60	4.60
1966	0.00	0.00	0.00	0.00
1967	0.00	0.00	1.24	1.24
1968	0.00	0.00	1.59	1.59
1969	0.00	0.00	6.75	6.75
1951–1969	179.10	2,898.57	1,981.71	5,059.38
1970	4.60	153.31	1.53	159.44
1971	0.00	0.00	18.70	18.70
1972	0.00	0.00	0.00	0.00
1973	0.00	0.00	6.67	6.67
1974	0.00	0.00	0.00	0.00
1975	0.00	0.00	5.52	5.52
1976	0.00	0.00	0.00	0.00
1977	1.53	0.00	3.07	4.60
1978	0.00	0.00	4.30	4.30
1979	0.00	0.00	0.00	0.00

TABLE 10. SUMMARY OF PROCESS GAS RELEASES AT ORGDP* (cont.)

YEAR	KG. OF PROCESS GAS RELEASED			
	K–29,K–31 K–33	K–1131	ALL OTHERS	TOTAL
1980	0.05	0.00	7.54	7.59
1981	2.00	0.00	2.01	4.01
1982	0.00	0.00	0.01	0.01
1983	0.00	0.00	0.00	0.00
1984	0.00	0.00	0.01	0.01
1985	0.00	0.00	0.07	0.07
1970–1985	8.18	153.31	49.43	210.92
1951 – 1985	187.28	3,051.88	2,031.14	5,270.30

* A. J. Legeay, Uranium Material Releases, May 21, 1982
R. T. Murr, Informal Communication, Accidental Releases of UF6 for the Period 1980 to Date, November 27, 1991

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Crawford

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Barlow

and K-27) from 1951 through 1985, when operations were suspended. Seven releases of 1 kg or greater are shown for the K-29, K-31 and K-33 buildings combined, of which 179 kg were released pre-1970 and 8 kg occurred in 1970 or later. Eleven large releases are shown for K-1131, the original feed facility, with 2,899 kg lost before 1970 and 153 kg lost after 1970. Releases from other facilities on the site are grouped into the last column labeled "All Others" and total 1,982 kg of UF_6 released before 1970 and 49 kg after 1970. As can be seen, most of the large process gas releases totalling 96% occurred during the early years before 1970. These results are shown graphically in Figure 12. Cleanup of released material was undertaken in each case. In many cases, total cleanup was not possible because of dispersion of some uranium over large distances.

Table 11 shows similar release information for the Paducah GDP between 1951 and 1991. Nine sizeable releases are shown to have occurred within process buildings between 1952 and 1969. A 3,078 kg release occurred at the C-333 vaporizer facility in 1960, and a fire in a C-337 cell in 1962 resulted in the release of 1,534 kg. For the four process buildings, 5,165 kg of releases occurred before 1970 but only 19 kg were lost after 1970. Eight sizeable process gas releases at the C-410 feed plant totaling 911 kg occurred between 1953 and 1969, but only one release of greater than 1 kg was lost thereafter. Releases at all other facilities totaled 3,018 kg between 1952 and 1969 and only a total of 49 kg since 1970. Thus, the bulk of the major releases, over 99%, occurred in the 1950s and early 1960s. Figure 13 shows a graphical presentation of these results.

The process gas releases at Portsmouth are summarized in Table 12. For the two major process buildings, X-330 and X-333, releases before 1970 totaled 2,874 kg and 256 kg after 1970. The feed facility, X-344, had releases of 1,271 kg before 1970 and a large loss of 6,014 kg after 1970. However, a major part of this was a single incident in 1978 involving 5,926 kg when a cylinder of liquid UF_6 was dropped in an outdoor storage yard and the cylinder wall ruptured, resulting in a loss of much of its content. This incident did occur in an outside yard, however, so no contamination was spread inside the X-344 building. Excluding this incident, the remaining losses from X-344 after 1970 totaled only 88 kg. For the combined X-344 building and all other buildings, the releases before 1970 totaled 1,544 kg or 88%, while those after 1970 totaled 205 kg or 12%. The releases for the Portsmouth GDP are shown plotted in Figure 14.

As demonstrated by the data for the three diffusion plants, the quantity and frequency of releases have decreased significantly over the nearly 40 years of operation with improvements in procedures and equipment, and experience. Consequently, contamination levels within all the facilities where UF_6 has been processed should be no greater than and in many cases lower than in the early days as cleanup of hot spots is performed in keeping with current practice on contamination control.

Another document, *Historical Radionuclide Releases From Current DOE Oak Ridge Operations Office Facilities*, OR-890, May 1988, contains much the same type of information as shown in Tables 10, 11, and 12. Some discrepancy, however, exists between the data in ORO-890 and Tables 10, 11, and 12. Since the information in these tables was derived from detailed lists of releases supplied by each of the GDPs, it was used for this analysis. No attempt was made to resolve the discrepancies at this time. The ORO report also includes vented release data from process facilities.

FIGURE 12. PROCESS GAS RELEASES AT ORGDP K-29,-31,-33 & ALL OTHERS

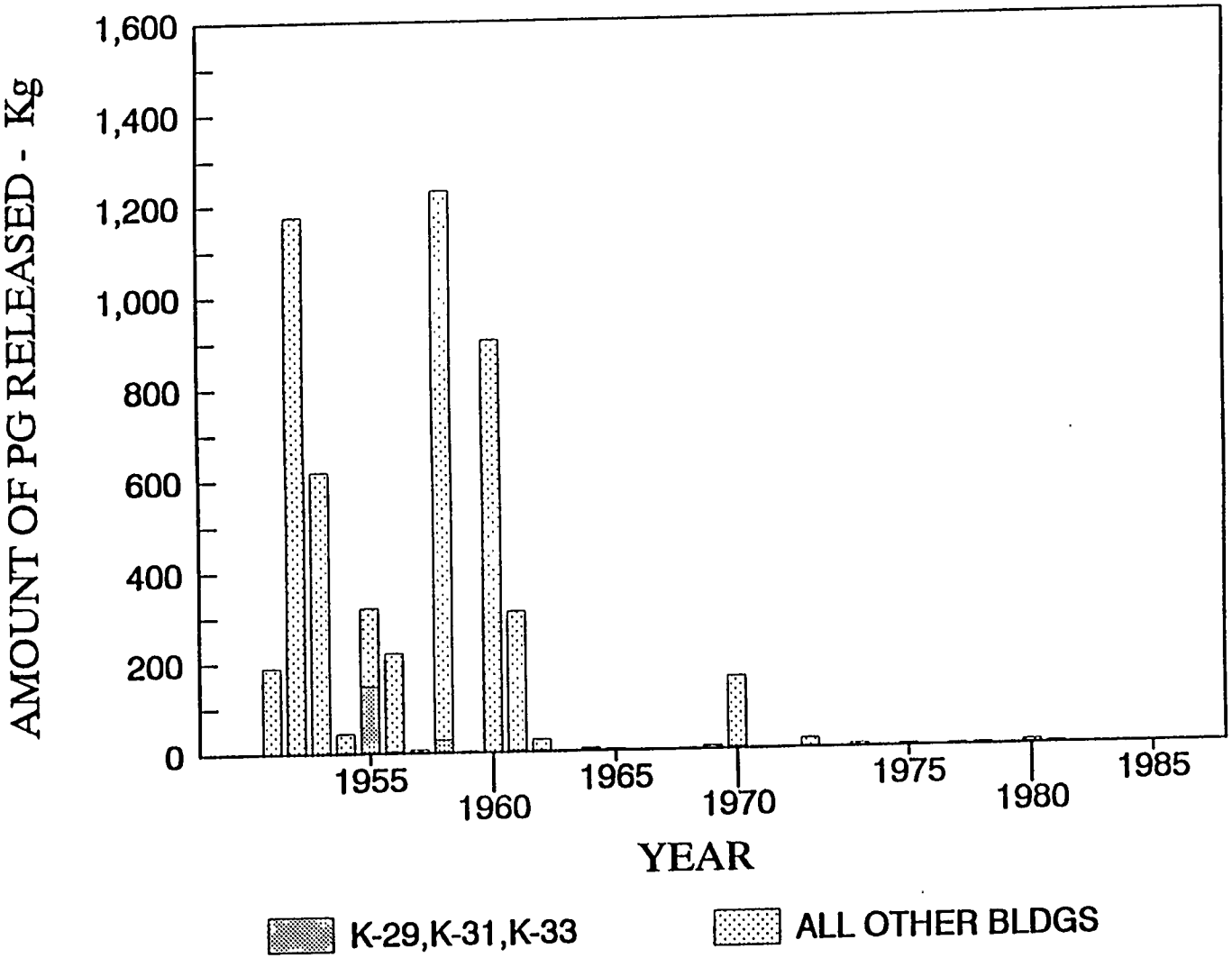


TABLE 11. SUMMARY OF PROCESS GAS RELEASES AT PGDP*

YEAR	KG. OF PROCESS GAS RELEASED			
	C–31,C–33 C–35,C–37	C–410	ALL OTHERS	TOTAL
1952	92.02	0.00	3.07	95.09
1953	57.67	162.41	507.92	728.00
1954	383.34	32.19	718.23	1,133.76
1955	0.00	346.52	432.54	779.06
1956	0.00	58.28	433.14	491.42
1957	3.07	11.34	50.46	64.87
1958	0.00	206.25	171.32	377.57
1959	9.20	89.45	63.37	162.02
1960	3,078.38	5.00	8.90	3,092.28
1961	0.00	0.00	30.28	30.28
1962	1,533.56	0.00	590.25	2,123.81
1963	0.00	0.00	0.00	0.00
1964	0.00	0.00	0.00	0.00
1965	0.00	0.00	0.00	0.00
1966	0.00	0.00	8.28	8.28
1967	0.00	0.00	0.00	0.00
1968	7.36	0.00	0.00	7.36
1969	0.00	0.00	0.00	0.00
1952–1969	5,164.60	911.44	3,017.76	9,093.80
1970	0.00	0.00	0.00	0.00
1971	14.72	0.00	0.00	14.72
1972	0.00	0.00	0.00	0.00
1973	0.00	0.00	0.00	0.00
1974	0.00	0.00	0.00	0.00
1975	0.00	0.00	22.70	22.70
1976	0.00	0.00	7.97	7.97
1977	0.00	0.00	0.00	0.00
1978	0.00	0.00	9.20	9.20
1979	0.00	0.00	3.07	3.07

TABLE 11. SUMMARY OF PROCESS GAS RELEASES AT PGDP*(cont.)

YEAR	KG. OF PROCESS GAS RELEASED			
	C-31,C-33 C-35,C-37	C-410	ALL OTHERS	TOTAL
1980	0.20	0.00	0.00	0.20
1981	0.00	0.00	0.04	0.04
1982	0.00	1.00	0.45	1.45
1983	0.00	0.00	1.54	1.54
1984	4.54	0.00	0.00	4.54
1985	0.02	0.00	2.80	2.82
1986	0.00	0.00	0.45	0.45
1987	0.00	0.00	0.00	0.00
1988	0.00	0.00	0.67	0.67
1989	0.00	0.00	0.50	0.50
1990	0.00	0.00	0.00	0.00
1991	0.00	0.00	0.00	0.00
1970-1991	19.48	1.00	49.39	69.87
1952-1991	5,184.08	912.44	3,067.15	9,163.67

* J. Dew, Initial Review of Available Radioactive Contamination Survey Data, October 23, 1991

D. A. Gill, PGDP UF6 Release Monitoring Program, Oct. 1, 1991

FIGURE 13. PROCESS GAS RELEASES AT PADUCAH PROCESS BLDGS & ALL OTHERS

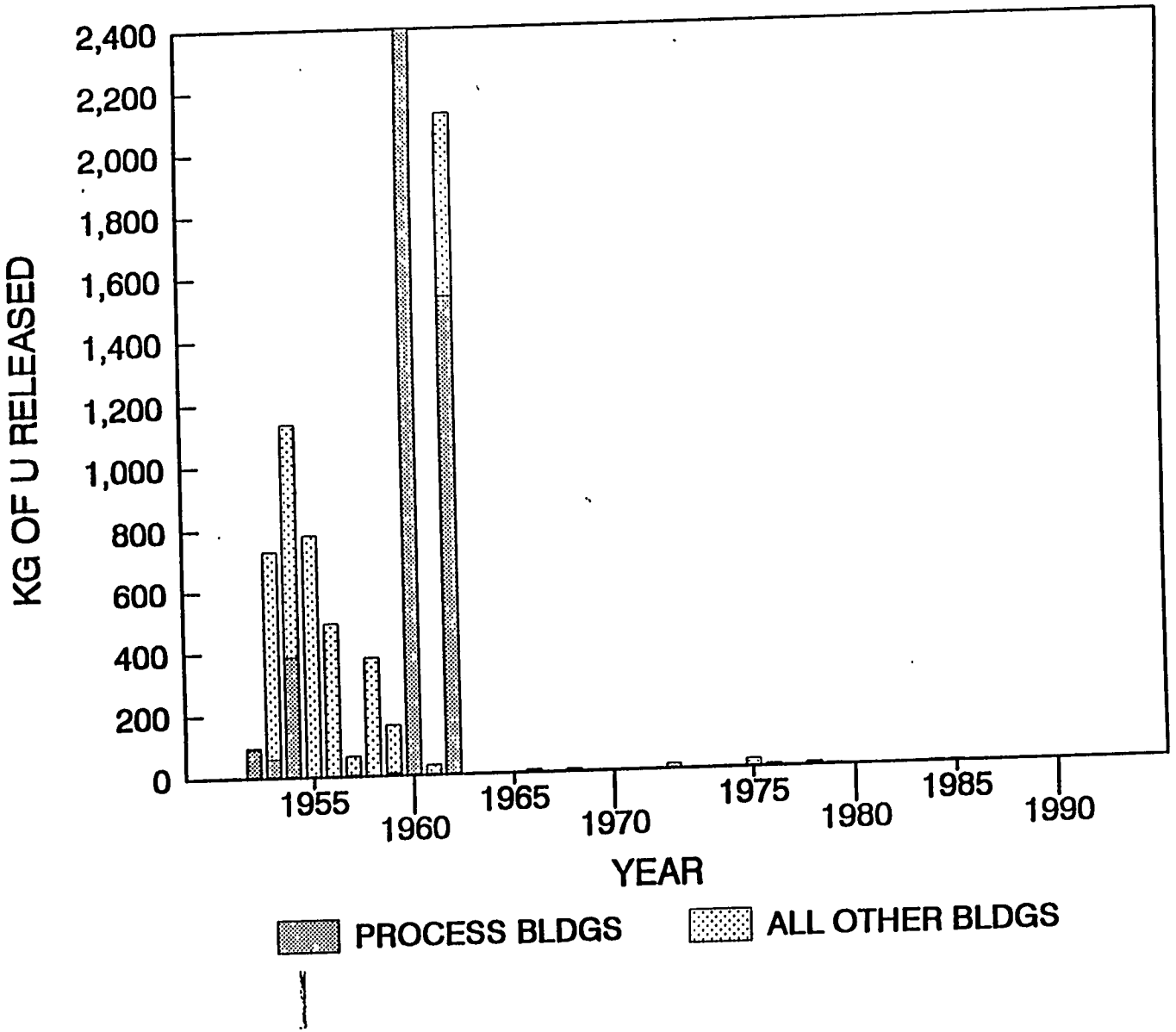


TABLE 12. SUMMARY OF PROCESS GAS RELEASES AT PORTSMOUTH GDP*

YEAR	KG. OF PROCESS GAS RELEASED			
	X-330, X-333	X-344	ALL OTHERS (exc. X-326)	TOTAL
1954	314.62	0.00	9.51	324.13
1955	1,878.52	0.00	99.57	1,978.09
1956	36.50	0.00	73.85	110.35
1957	22.00	0.00	5.92	27.92
1958	21.84	600.29	1.92	624.05
1959	0.00	68.24	38.36	106.60
1960	138.90	406.25	6.67	551.82
1961	1.40	194.95	13.50	209.85
1962	0.00	1.53	6.58	8.11
1963	0.00	0.00	2.96	2.96
1964	0.00	0.00	0.00	0.00
1965	0.00	0.00	14.00	14.00
1966	0.00	0.00	0.00	0.00
1967	0.00	0.00	0.00	0.00
1968	0.00	0.00	0.00	0.00
1969	460.00		0.00	460.00
1954-1969	2,873.78	1,271.26	272.84	4,417.88
1970	0.00	1.00	18.30	19.30
1971	28.00	0.00	0.00	28.00
1972	0.00	0.00	13.20	13.20
1973	0.00	0.00	59.40	59.40
1974	0.00	0.00	0.00	0.00
1975	0.00	10.59	0.00	10.59
1976	0.00	64.50	0.00	64.50
1977	50.00	5.58	7.97	63.55
1978	102.40	5,925.80 **	5.83	6,034.03
1979	3.00	0.00	0.00	3.00

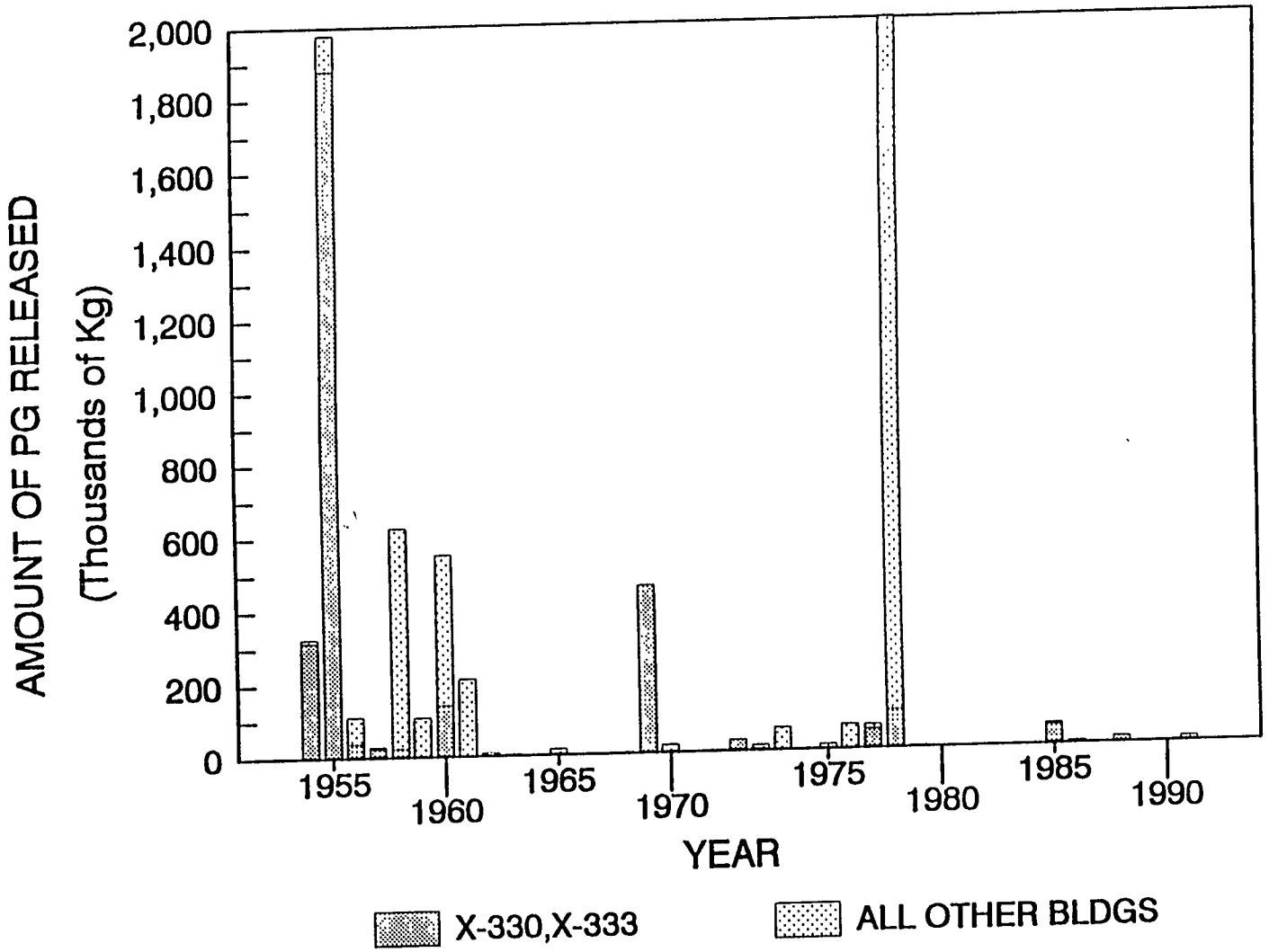
TABLE 12. SUMMARY OF PROCESS GAS RELEASES AT PORTSMOUTH GDP*(cont.)

YEAR	KG. OF PROCESS GAS RELEASED			
	X-330, X-333	X-344	ALL OTHERS (exc. X-326)	TOTAL
1980	0.00	0.00	0.00	0.00
1981	1.07	0.00	0.00	1.07
1982	0.00	0.00	0.00	0.00
1983	1.90	0.00	0.00	1.90
1984	0.00	0.00	0.00	0.00
1985	49.48	6.47	0.00	55.96
1986	6.35	0.00	0.00	6.35
1987	0.00	0.00	0.00	0.00
1988	13.75	0.00	0.00	13.75
1989	0.00	0.00	0.00	0.00
1990	0.00	0.00	0.00	0.00
1991	0.00	0.00	11.98	11.98
1970-1991	255.95	6,013.94	116.68	6,386.58
1954-1991	3,129.74	7,285.20	389.52	10,804.45

* D. A. Shisler, Portsmouth Gaseous Diffusion Plant Historical Release Data, November 13, 1991

** Cylinder ruptured in outdoor storage yard during handling.

FIGURE 14. PROCESS GAS RELEASES AT PORTSMOUTH PROCESS BLDGS & ALL OTHERS



CONTAMINATION WITHIN THE PROCESS BUILDINGS

Because of the many incidents of accidental process gas releases, contamination of the interior of process buildings with uranium and its compounds would appear to be an obvious conclusion. Records of building surveys conducted in the early days of operations have not yet been located, if they exist at all. As a result, the only information on contamination levels existing in process buildings and in the many auxiliary buildings in which UF_6 is handled either as a gas or a liquid is in relatively recent surveys conducted at each of the three diffusion plant sites. The levels of contamination found in these surveys are considered to represent a relatively constant level over the years because contamination control practice dictates that heavily contaminated areas be decontaminated to a lower level so that all areas in process buildings are available for routine access.

At the request of J. W. Bennett in a memorandum dated July 17, 1991, surveys of process buildings were performed in each of the process buildings at Oak Ridge, Paducah and Portsmouth. These were relatively limited surveys performed in a very short period to generate data on contamination of floors, walls and columns of cell floors and operating floors. Criteria used as specified in the J. W. Bennett letter were taken from NRC Regulation Guide 1.86 and were given as 5,000 dpm/100 cm^2 averaged over one square meter for alpha or beta-gamma total (fixed and removable) radiation and 1,000 dpm/100 cm^2 averaged over one square meter for alpha or beta-gamma for transferable radiation.

At Oak Ridge, results of the special survey performed to meet the request were reported in a letter by L. E. Hall[15]. These results are summarized in Table 13 for all five process buildings. Because of time constraints, a single unit in each building was chosen to be surveyed as representative of the entire building. The results for K-25 and K-27, which are high-assay buildings and thus directly related to production for defense, are included because those buildings were shut down in 1964. The levels existing today in these buildings are believed to be representative of the contamination existing in all of the cascade buildings during the 1960s when enrichment was being performed solely for defense purposes. Compared to the extent of contamination found in Buildings K-25 and K-27, the contamination found in K-31, which was operated 20 years longer, is considered to about the same level, while the extent of contamination found in K-33 is definitely lower.

The extent of contamination found in K-31 and K-33 indicates that even though these buildings were operated until 1985 and throughout a major equipment exchange program (the CIP/CUP program conducted in 1975-1982), their contamination is either lower or no greater than that of buildings that were operated only through 1964. Enrichment operations through the mid-sixties were solely for defense purposes. The extent of contamination currently existing in K-31 and K-33 should be ascribed to contamination incurred in the early days and to the first Improvement Program; contamination probably has stayed relatively constant since.

The extent of contamination for K-29 is obviously much higher on both the operating (first) floor and on the cell (second) floor. This is probably the result of an incident in 1981 in which a large quantity of process gas was released following an exothermic reaction in Unit 2 of this building. Since no cell

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15. L. E. Hall, Letter to Bryan Walker, "Characterizing Process Building Contamination - Contamination Estimate," July 31, 1991

TABLE 13. ESTIMATE OF CASCADE BUILDING CONTAMINATION – OAK RIDGE

LOCATION	PERCENT AREA CONTAMINATED – LEU CASCADE		
	K–29 Unit 3	K–31 Unit 3	K–33 Unit 5
Cell Floor			
Floor	100%	15%	5%
Walls	60%	10%	5%
Columns/Beams	95%	10%	5%
Operating Floor			
Floor	70%	15%	5%
Walls	95%	10%	5%
Columns/Beams	90%	10%	5%

LOCATION	PERCENT AREA CONTAMINATED – HEU CASCADE		
	K–25 306–6	K–27 Unit 7	
Cell Floor			
Floor	15%	20%	
Walls	10%	25%	
Columns/Beams	10%	35%	
Operating Floor			
Floor	15%	10%	
Walls	10%	15%	
Columns/Beams	10%	50%	

housings enclose the process equipment in this building, the resulting UO_2F_2 was widely distributed throughout both cell and operating floors of the building. Thus, the amount of contamination found in K-29 is considered to be atypical of other process buildings.

The Paducah plant's response[16] for characterization of its process buildings is shown in Table 14. Paducah selected 30 to 50 grid areas in each of the four process buildings, C-31, C-33, C-35 and C-37 and randomly sampled five points in each grid area for both direct beta-gamma and alpha activity. Internal cell housings and immediately adjacent floor areas were not surveyed because these areas were known to have levels of contamination above specified limits.[17] The survey revealed that large percentages of floor areas in all of the process buildings had measurable amounts of contamination, primarily beta-gamma rather than alpha. Contamination levels on the ground floor (operating floor) were found to be higher than those on the cell floor (second floor), and this fact was ascribed to the past practice of temporarily storing process equipment that was in the process of being transferred from the process building to the decontamination facility in preparation for repair or upgrading. Also, quantities of green salt (UF_4) in drums were often stored for extended periods at ground levels. Another reason given was the probability of wash down from leaking recirculating water (RCW) systems into floor drains on the ground floor. The occasionally large areas of contamination on columns near the floor were attributed to contaminants being swept against the base of such columns.

Painting of columns from time to time has fixed the contamination at such locations.

The results of the contamination survey performed at Portsmouth were reported in a letter by R. G. Donnelly[18] and are shown in Table 15. In routine monitoring for beta surface contamination at Portsmouth, net beta-gamma count rate of 100 counts per minute (cpm) from a pancake probe in contact with a uniformly contaminated surface was assumed to be equivalent to 5,000 dpm/100 cm^2 based on the probe efficiency in detecting ^{99}Tc contamination[19]. This calibration was used in obtaining the area percentages shown in Table 15. However, in the X-333 and X-330 buildings, which are low-assay buildings, beta contamination is dominated (in most cases) by ^{238}U daughter emissions. The count rate equivalent to 5,000 dpm /100 cm^2 for uranium decay product contamination is generally considered to be about 170 cpm. This results in much lower area percentages as also shown in Table 15, and these values of 11% in X-330 and 18% in X-333 are considered to be

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16. S. A. Polston, Letter to D. C. Booher, "Characterizing Process Building Contamination", July 29, 1991

17. The limits as specified in J. W. Bennett's letter are from NRC Regulation Guide 1.86 and represent limits beyond which contamination control measures must be taken in accordance with current policy. Consequently, no health hazards are considered to exist when proper procedures are followed.

18. R. G. Donnelly, Letter to E. W. Gillespie, "Characterizing Process Building Contamination - Contamination Estimate," July 29, 1991

19. Ibid

TABLE 14. ESTIMATE OF CASCADE BUILDING CONTAMINATION – PADUCAH

LOCATION	PERCENT OF AREA CONTAMINATED			
	C–331	C–335	C–333	C–337
Cell Floor				
Floor	50%	30%	55%	78%
Walls	4%	12%	0%	29%
Columns/Beams	50%	56%	15%	24%
Operating Floor				
Floor	77%	90%	68%	43%
Walls	0%	0%	0%	7%
Columns/Beams	8%	0%	10%	7%

TABLE 15. ESTIMATE OF CASCADE BUILDING CONTAMINATION – PORTSMOUTH

LOCATION	PERCENT AREA CONTAMINATED – LEU CASCADE			
	Beta–Gamma 100 cpm*		Beta–Gamma 170 cpm*	
	X–330	X–333	X–330	X–333
Cell and Operating Levels	38%	47%	11%	18%
Floor	2%	1%		
Walls	5%	10%		
Columns/Beams				

LOCATION	PERCENT AREA CONTAMINATED – HEU CASCADE		
		X–326	
Cell and Operating Levels		29%	
Floor		16%	
Walls		19%	
Columns/Beams			

*Probe efficiency based on technetium (100 cpm) or uranium decay products (170 cpm)

more representative of the actual contamination existing in LEU buildings[20]. Thus, contamination levels in X-330 and X-333 buildings are lower than in the X-326 building.

The fact that contamination at Portsmouth appears to be lower than at Paducah may be ascribed to several factors, among them (1) the counting technique as mentioned previously, (2) storage of equipment and green salt at Paducah, and (3) procedural differences in the manner in which the surveys were conducted over a very short period.

The widespread contamination in all process buildings at Oak Ridge, Paducah and Portsmouth has occurred because of accidental process gas releases and because of numerous equipment changeouts that have been performed since the beginning of operations of the plant.

Oak Ridge Survey Results, 1982-1985

A very comprehensive survey of contamination in process buildings at Oak Ridge was performed in 1982-1984 based primarily on alpha readings[21]. All of the process buildings were surveyed in a very thorough fashion, with 4,400 readings taken in K-29, 11,300 readings in K-31, and 23,900 readings in K-33. Also, 15,300 readings were taken in K-27, and over 73,700 readings were taken in K-25. DOE Order 5480.11 places 4,000 to 20,000 dpm/100 cm^2 in Class II category of contamination and greater than 20,000 dpm/100 cm^2 in Class III, both of which are considered as regulated zones. Thus, all contaminated areas with Class II and Class III levels of contaminations fall in the range of contamination of 5,000 dpm/100 cm^2 or greater. Table 16 is a summary of the Mahathy and Ward data as presented in the Characterization Reports on K-29[22], K-31[23], K-33[24], K-25[25] and K-27[26].

Table 16 shows that based on DOE Order 5480.11 guidelines for Classes II and III, contaminated areas on cell floors in LEU buildings, K-29, K-31 and K-33, range from 0.4% to 1.3%, while on cell

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20. B. S. Manninen, Informal communication, December 18, 1991

21. J. M. Mahathy and M. E. Ward, *ORGDP Building Radiation Background Quantification: 1982-1985*, August 17, 1985 (K/HS-82).

22. W. S. Lenihan, et.al., *Building Characterization Report, ORGDP Permanent Shutdown, Building K-29*, April 29, 1988 (K/D-5750).

23. J. M. Chiang, et.al., *Building Characterization Report, ORGDP Permanent Shutdown, Building K-31*, April 19, 1988 (K/D-5751).

24. J. M. Chiang, et.al., *Building Characterization Report, ORGDP Permanent Shutdown, Building K-33*, April 6, 1988 (K/D-5752).

25. T. Shapiro, et.al., *Building Characterization Report, ORGDP Permanent Shutdown, Building K-25*, March 10, 1988 (K/D-5748).

26. W. S. Lenihan, et.al., *Building Characterization Report, ORGDP Permanent Shutdown, K-27*, March 22, 1988 (K/D-5749).

TABLE 16. ESTIMATE OF CASCADE BUILDING CONTAMINATION – OAK RIDGE

ALPHA READINGS ONLY*

BASED ON DOE 5480.11 GUIDELINES

LOCATION	LEU CASCADE			HEU CASCADE	
	K–29	K–31	K–33	K–25	K–27
Cell Floor	1.3%	1.1%	0.4%	0.9%	2.6%
Operating Floor	0.2%	0.2%	0.3%	0.7%	0.6%
Cells	0.9%	0.4%	0.0%	2.7%	5.5%

* J. M. Mahathy and M. E. Ward, "ORGDP Building Radiation Background Quantification: 1982–1985, August 16, 1985 (K/HS–82)

floors in HEU buildings, K-25 and K-27, contaminated areas range from 0.9% to 2.6%. On operating floors, contaminated areas range from 0.2% to 0.3% in LEU buildings and from 0.6% to 0.7% in HEU buildings. Survey data from some individual cells are also included in Table 16. These surveys were made of the cell floor area in the immediate vicinity of some cells in each of the buildings and as such are partial duplications of the cell-floor surveys, which covered all of the cell-floor area. Cell information for LEU buildings shows contamination ranging from 0% to 0.9%, while for HEU buildings, the area percentages are 2.7% to 5.5%.

The information in Table 16 **based on alpha readings only** would indicate that percentages of contaminated areas are equal to or slightly lower in K-29, K-31 and K-33 than in corresponding locations in K-25 and K-27. This fact would indicate that the additional years of operation of LEU buildings did little, if any, to increase the extent of contamination in these buildings. Based on this information, the level of decontamination effort required for process buildings is no greater now than the effort that would have been required in 1965 if all the facilities had been required to have been decommissioned at that time. (Note: The specific activity of 93% enriched uranium is approximately 100 times that of natural uranium, based upon the increased concentration of ^{234}U in the product stream. The specific activity of 5% enriched uranium is 3.5 times that of natural uranium. Consequently, a smaller quantity of enriched material would be required to exhibit the same level of activity as normal or slightly enriched uranium[27]).

URINALYSIS FOR PERSONNEL EXPOSURE TO CONTAMINATION

Exposure of the GDP work force to radioactive contamination has been a matter of concern since the initiation of the uranium enrichment process. While alpha emission of uranium is not considered an external hazard, ingestion of uranium particles is of concern. Consequently, a bioassay program of urinalyses of workers who were exposed to contaminated working conditions was instituted from the very beginning of enrichment operations. Much of this information has been placed into a computerized database at Oak Ridge and Portsmouth from which it can be retrieved for analysis. While information available does not pinpoint the nature of workers' activities or their work stations, the data are an indication of the times at which uranium contamination or releases were occurring. Since uranium particles are ingested primarily by breathing in contaminated areas or near contaminated equipment, a study of urinalyses can be expected to provide a guide to the occurrence of contamination in work areas.

The number of workers exposed to uranium contamination will vary with the nature of activities occurring in the diffusion plants. During periods when numerous equipment failures occurred, and particularly when an equipment exchange/upgrading program was under way, the number of exposed workers rose significantly. Thus, the number of urine samples revealing significant levels of uranium or alpha activity is expected to confirm the extent of uranium contamination existing in the workplace during such periods of increased activity. Urinalysis data are available for Oak Ridge and Portsmouth plants from the early days of operation through the present, thus allowing an estimate of the extent of contamination existing in the work areas over almost the entire period of operation of these plants. Urinalysis data for Paducah were also taken but have not been entered into a computerized data base, so they were not examined for this study.

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27. J. C. Bailey, letter to H. M. Noritake, January 14, 1992

The urinalysis data for ORGDP personnel are presented in Table 17. Two types of contamination readings were chosen to be displayed, the number of samples giving alpha readings above 6 dpm/100 ml and number of samples with uranium levels greater than 0.017 mg/l. Figure 15 is a graph showing the number of samples with alpha readings above 6 dpm/100 ml. A very large number of high alpha readings were observed in the 1953 to 1966 time frame. The period from 1953 to 1957 may be considered the start-up period for the K-31 and K-33 buildings when lack of experience in the work force would be a major problem leading to errors in operation and accidental releases of process gas. The remainder of the period from 1957 to 1962 was a period of extensive equipment changeout as the first cascade improvement involved a large number of stages in both the axial and centrifugal portions of the cascade. Thus, numerous opportunities occurred for plant personnel to be exposed to uranium contamination as indicated by the data.

Another increase in alpha counts occurred in the 1975 to 1983 time frame, which corresponds to the CIP/CUP period. The number of samples testing high was lower than in the earlier period, however, indicating possibly that (a) protective measures were imposed, (b) work areas had less free-floating contamination, or (c) equipment was less contaminated than previously experienced. Each of these factors probably contributed to a portion of the improvement. While the available data do not permit confirmation of these assumptions, apparently better controls on contamination levels in the work place were in place in the later program.

Figure 16 shows a display of uranium readings and exhibits the same pattern of higher numbers of contaminated samples in the 1953 to 1963 time frame and again in the 1975 to 1982 time frame, corresponding to the two major equipment replacement programs. Thus, contamination of process buildings and other facilities that handled process equipment took place starting from very early in the operation of the enrichment plants, and much of this contamination probably remains in place today as fixed contamination.

Urinalysis data for the Portsmouth GDP is listed in Table 18. The standards used in the selection of data for Portsmouth are alpha readings above 8 dpm/100 ml and uranium measurements above 0.02 mg/l or slightly higher standards than for Oak Ridge. The number of samples with alpha readings greater than 8 dpm/100 ml is plotted in Figure 17. The results are somewhat higher than at Oak Ridge during the early years from 1956 through 1961 and at irregular intervals following the early improvement program. No significant increase in the number of samples exceeding the target value is seen for the period of the CIP/CUP program, however.

The number of samples exceeding the uranium target of 0.02 mg/l is plotted in Figure 18. The largest number of samples exceeding the 0.02 mg/l occurs during the 1955-1961 improvement program as with the alpha readings. After 1961, however, the number of samples exceeding the target falls off very sharply and remains low thereafter with the single exception of 1978 when a liquid cylinder was dropped, resulting in a rupture and major release of process gas.

While the urinalysis data cannot be considered as being conclusive in confirming the existence of contaminated work areas, it is another indicator that work areas were contaminated very early in the operation of the enrichment plants. With improvements in operating procedures as well as in contamination control procedures, the presence of uranium in urine samples of the work force has been reduced significantly over the years.

TABLE 17. ORGDP URINALYSIS DATA, 1948 – 1985*

YEAR	NO. OF SAMPLES WITH	
	ALPHA READINGS >6 dpm/100 ml	URANIUM READINGS >0.017 mg/l
1948	8	79
1949	6	63
1950	9	96
1951	14	164
1952	154	534
1953	163	493
1954	107	359
1955	166	324
1956	171	468
1957	220	429
1958	198	323
1959	163	561
1960	234	449
1961	235	343
1962	161	120
1963	131	59
1964	87	53
1965	116	55
1966	39	50
1967	21	42
1968	37	43
1969	29	35
1970	26	52
1971	53	64
1972	38	47
1973	62	88
1974	19	33

TABLE 17. ORGDP URINALYSIS DATA, 1948 – 1985* (cont.)

YEAR	NO. OF SAMPLES WITH	
	ALPHA READINGS >6 dpm/100 ml	URANIUM READINGS >0.017 mg/l
1975	65	144
1976	56	171
1977	123	542
1978	98	446
1979	78	237
1980	81	194
1981	51	106
1982	19	29
1983	15	42
1984	44	60
1985	40	60

* J. N. Ingle and W. G. Tankersley, ORAU, Informal Communication, October 29, 1991

FIGURE 15. URINALYSIS DATA - OAK RIDGE ALPHA READINGS ABOVE 6 dpm/100 ml

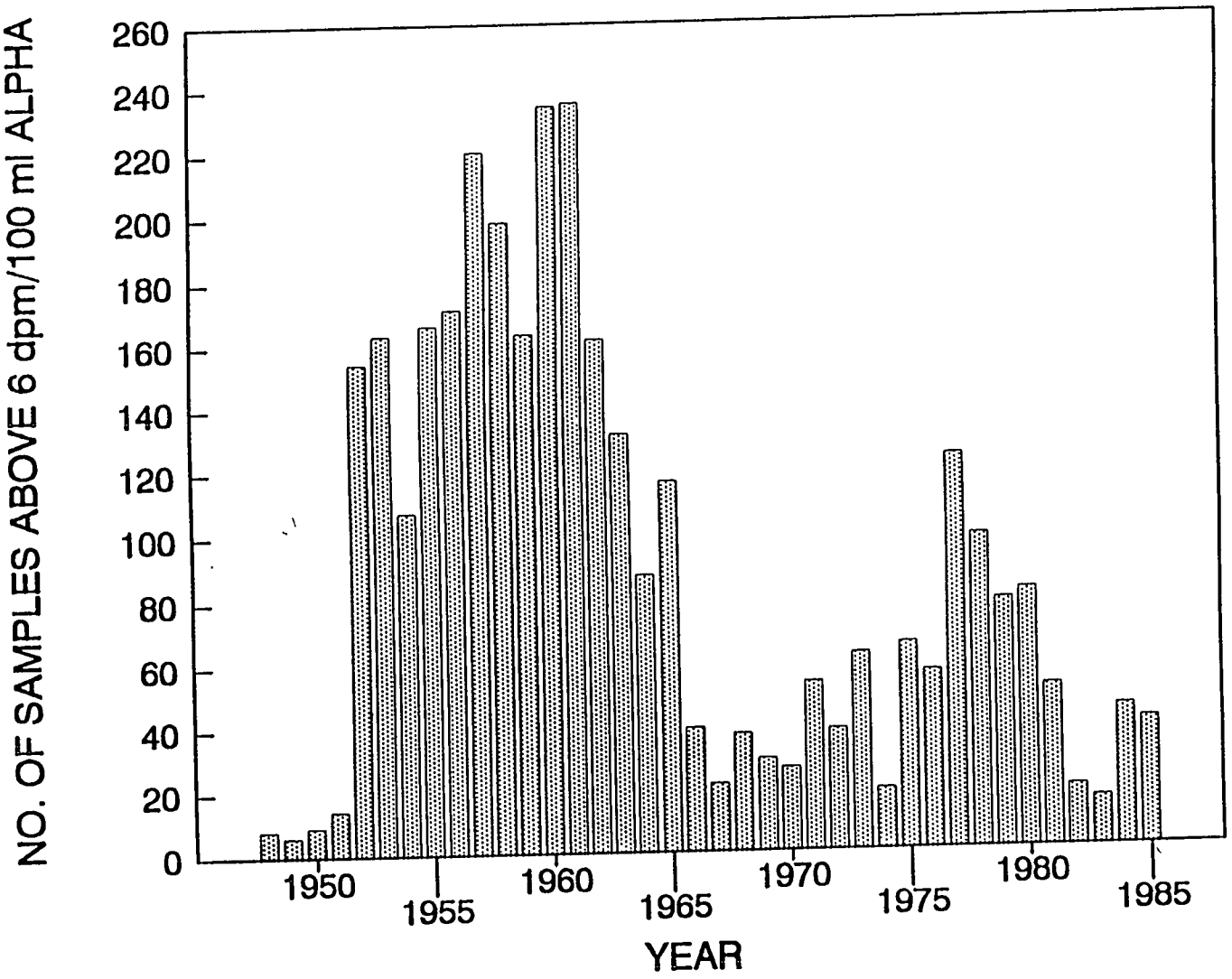


FIGURE 16. URINALYSIS DATA - ORDGP
URANIUM READINGS ABOVE 0.017 mg/l

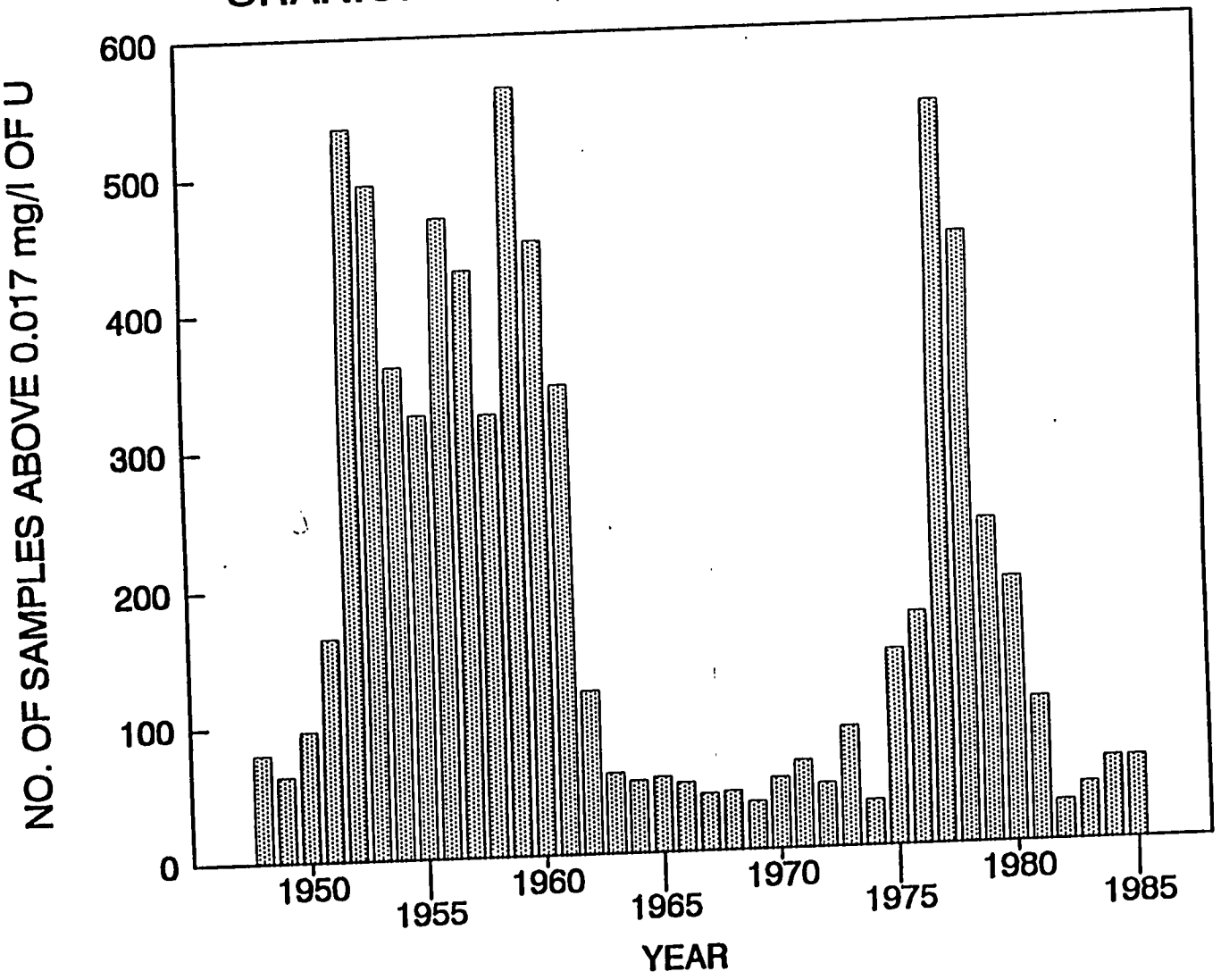


TABLE 18. PORTSMOUTH URINALYSIS DATA, 1955 – 1990*

YEAR	NO. OF SAMPLES WITH	
	ALPHA READINGS > 8 dpm/100 ml	URANIUM READINGS > 0.02 mg/l
1955	31	470
1956	251	128
1957	174	194
1958	206	252
1959	301	252
1960	354	122
1961	256	152
1962	46	21
1963	57	13
1964	37	6
1965	208	36
1966	40	18
1967	33	15
1968	40	19
1969	224	16
1970	114	25
1971	129	23
1972	49	14
1973	243	34
1974	48	22
1975	223	59
1976	81	27
1977	80	57
1978	117	126
1979	49	55
1980	44	37
1981	64	29
1982	73	26
1983	39	7

TABLE 18. PORTSMOUTH URINALYSIS DATA, 1955 – 1990* (cont.)

YEAR	NO. OF SAMPLES WITH	
	ALPHA READINGS > 8 dpm/100 ml	URANIUM READINGS > 0.02 mg/l
1984	36	12
1985	46	41
1986	32	21
1987	107	22
1988	82	9
1989	49	17
1990		42

* E. R. Wagner, Portamouth, Informal Communication, November 5, 1991
C. R. Dulin, Portsmouth, Informal Communication, November 11, 1991

FIGURE 17. URINALYSIS DATA - PORTSMOUTH
ALPHA READINGS ABOVE 8 dpm/100 ml

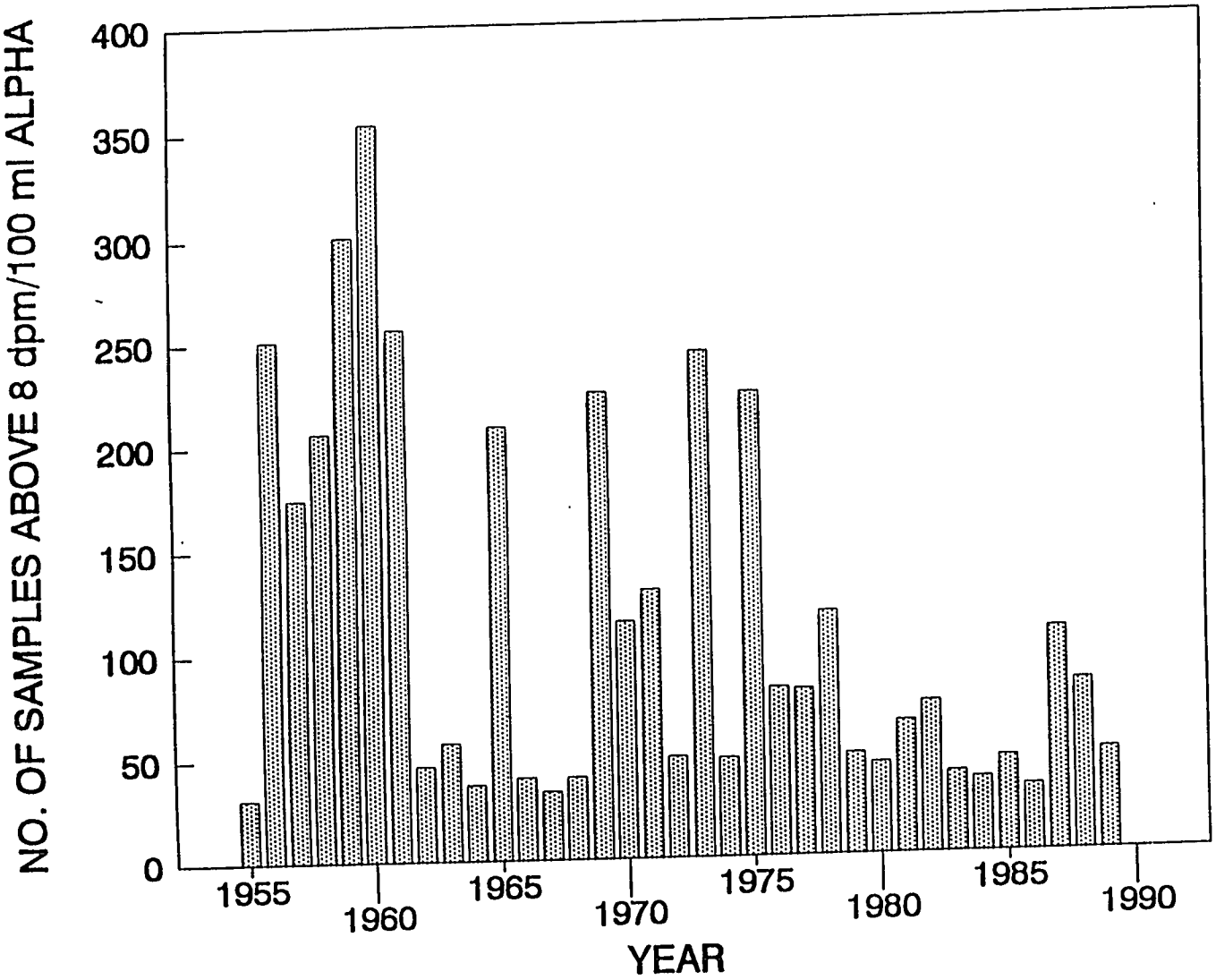
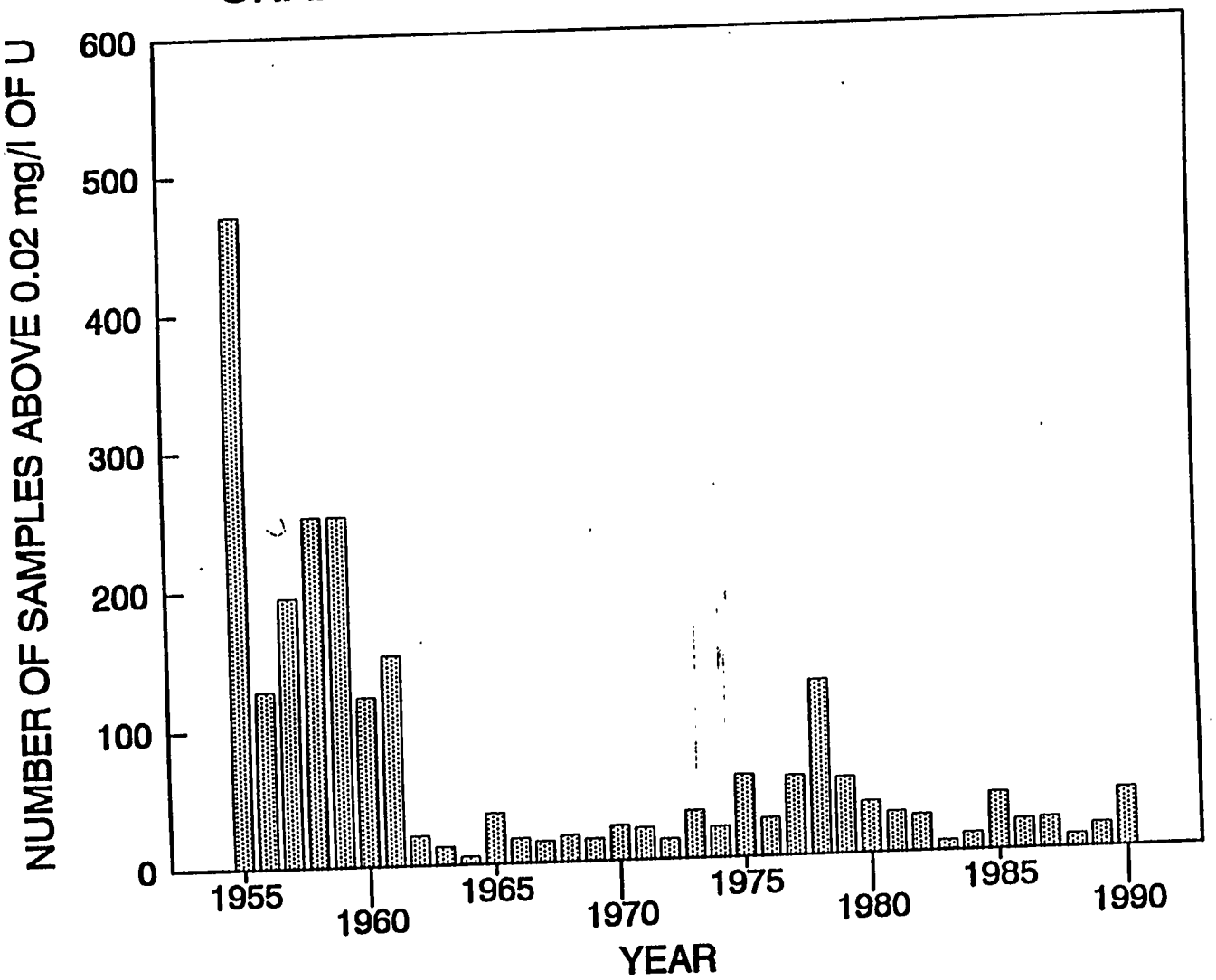


FIGURE 18. URINALYSIS DATA - PORTSMOUTH URANIUM READINGS ABOVE 0.02 mg/l



OBSERVATIONS AND CONCLUSIONS

Information presented in this report provides a database from which preliminary conclusions can be drawn regarding responsibility for the cost of D&D of the GDPs at Oak Ridge, Paducah and Portsmouth. The requirement for decontamination or disposal of process equipment as low-level waste because of **internal radioactive contamination** was established by early operation of the equipment for the defense mission.

- Internal contamination of process equipment occurs when process gas is first introduced because of immediate chemisorption of uranium fluorides.

- The NDA survey confirms the fact that equipment installed in the fifties and sixties contains greater uranium deposits than new equipment installed in the late seventies. This fact is also indirectly supported by the inventory difference data for each plant.

- Introduction of reprocessed uranium from defense production reactors into the gaseous diffusion process contaminated all three plants with trace amounts of transuranics and ^{99}Tc.

Many large buildings at the GDP sites have become contaminated with radioactivity during the many years of operation and represent a very large D&D cost category. **Building contamination** is dependent on sources of uranium released within the process buildings. The data for two principal mechanisms for release, accidental releases and equipment changeouts, show that the early operation of the facilities was characterized by frequent maintenance actions and significant accidental releases. Consequently, by the early 1960s, process buildings and other buildings were significantly contaminated.

- A large number of equipment exchanges to replace failed equipment or to install upgraded equipment have provided opportunities to spread contamination within process buildings. Many of these changeouts occurred before 1970, and contamination controls have been significantly enhanced since the early years of operation.

- Process gas releases are another source of contamination of buildings on the sites. A majority of these releases occurred during the 1950s and early 1960s.

- Surveys of contamination within process buildings have proved that (1) contamination exists to some extent in every process building; and (2) K-25 and K-27 buildings at Oak Ridge that were shut down in 1964 have contamination levels comparable to or higher than the low assay process buildings, K-31 or K-33 (K-29 was subjected to a major release) indicating that incremental contamination since 1965 has been quite small.

- Urinalysis samples of workers confirm the fact that many workers were exposed to uranium released in their work areas in the early years of operation and during programs for equipment exchanges.

Based on this information, the level of decontamination effort required for process buildings is no greater now than the effort that would have been required in 1965 if all the facilities had been required to be decommissioned at that time.

While no single set of data provides a clear basis for allocation of D&D liability, the preponderance of all data reviewed clearly supports the position that a large percentage of D&D costs are the responsibility of the U. S. Government. The gaseous diffusion plants were originally constructed to fulfill the government's need for enriched uranium. Only after the first two decades of government use did the commercial sector derive any benefit from the existence of these plants. In 1969, when commercial production began, the diffusion plants were already contaminated and the need to conduct D&D was well established. Contamination with radioactivity at the three GDP sites consists of internal contamination of equipment and contamination of building interiors. As suggested by historical data gathered to date, the introduction of commercial production did not add greatly to the existing overall level of contamination. This is particularly true of internal contamination of equipment where some data suggest that the levels have actually been reduced during the years of commercial production. Furthermore, the large volume of contaminated material has remained relatively unchanged with only modest increases associated with process equipment upgrades.